50392

RF/ER 94-0027 UN

CHEMICALLY ENHANCED STEAM STRIPPING OF RADIONUCLIDES FROM RFETS SOILS TREATABILITY STUDY REPORT

INTERIM PHASE I REPORT

ADMIN RECORD

DOCUMENT CLASSIFICATION
REVIEW WAIVER PER
CLASSIFICATION OFFICE

U S DEPARTMENT OF ENERGY/ROCKY FLATS FIELD OFFICE
Rocky Flats Environmental Technology Site
Golden Colorado

ENVIRONMENTAL RESTORATION PROGRAM DIVISION

TABLE OF CONTENTS

TABLE OF ACRONYMS

ER	Environmental Restoration
INEL	Idaho National Engineering Laboratory
LANL	Los Alamos National Laboratories
NAPL	Non Aqueous Phase Liquid
OU2	Operable Unit 2
RF	Rocky Flats
RFETS	Rocky Flats Environmental Technology Site
VISITT	Vendor Information Service for Innovative Treatment Technologies
VOC	Volatile Organic Compound

ABSTRACT/EXECUTIVE SUMMARY

Treatability studies to remove Pu and Am from contaminated soils in Operable Unit 2 of the 903 Pad Area at the Rocky Flats Plant are being conducted The soil in Area 2 was contaminated as a result of drums that were stored in that area which leaked The remediation technology under development for the removal of actinides from contaminated soils uses a leaching scheme (involving redox and complexing agents) to chemically enhance the steam stripping process This report focuses on Phase I of the development effort which consists of selecting leaching schemes capable of removing Pu and Am from soils at the temperatures expected during the steam stripping process The selected leaching schemes would then be tested in conjunction with steam stripping to remove actinides from Rocky Flats soils at the bench scale

The techniques used in Phase I of this report to select the most promising leaching schemes consisted of batch desorption experiments and column studies Batch desorption experiments consisted of equilibrating contaminated soil with a leaching solution separating the phases and determining the amount of Am and Pu in each phase These batch desorption experiments were performed at 20 and 80 C Column experiments were used to corroborate the findings of the batch experiments after one of the most promising leaching schemes was selected

Using complexants alone at 20 C over 60% of the americium and 50% of the plutonium in the soil can be removed Combining complexants with a reducing or oxidizing agent yields the best results More than 70% of the americium and 70% of the plutonium in Rocky Flats soils can be removed at 20 C by using a complexant and a reducing agent At 20°C the largest amounts of Am were removed (between 70 80%) with those leaching schemes in which dithionite was used in a pH range of 5 4 7 4 with the following complexants citric acid NTA EGTA DTPA EDTA At 20 C the largest Pu removal (i e 75%) was accomplished with NTA / dithionite at a pH of ~5 5

For approximately 40% of the leaching schemes the removal of Am from Rocky Flats soils was found to increase with increasing temperature For approximately 60% of the leaching schemes there was no practical difference in the removal of Am with increasing temperature (i e ±10% difference) In only two leaching schemes (Na_3Citrate / 3% H_2O_2 and NTA / 3 33 eq NaOH / $Na_2S_2O_8$) was there a decrease in percent Am removal with increasing temperature The same trends were observed for Pu with the exception of the following two leaching schemes (Na_2EDTA/1 eq NaOH / 3% H_2O_2 and DTPA / 3 45 eq NaOH / 3% H_2O_2) in which the percent Pu removal decreased with an increase in temperature

At 80°C it is possible to remove over 70% of the Am in the soil and 60% of the Pu in the soil without the utilization of a redox agent The use of complexants with redox agents at 80 C removed over 80% of the Am in the soil and over 70% of the Pu from the soil At 80 C the largest amounts of Am were removed (between 80 90%) with those leaching schemes in which dithionite was used in a pH range of 5 4 7 4 with the following complexants NTA EGTA DTPA EDTA At 80 C the largest Pu removal (i e 78%) was accomplished with NTA / dithionite at a pH of ~5 5

Column experiments indicated that over 70% of the alpha activity could be removed from soil columns at 20 and 75 C by using sodium citrate in conjunction with a reducing agent such as ascorbic acid or dithionite These results corroborate those found with the batch desorption experiments

1 0 INTRODUCTION

1 1 Site History

The particular focus of this study is Operational Unit 2 903 Pad area which potentially contains plutonium americium and organic solvents Experiments were performed on soils which were collected from the 903 Pad by Rocky Flats personnel The 903 Pad Area encompassing the original 903 Drum Storage Site was used from October 1958 to January 1967 for storage of radioactive contaminated oil drums whose contents were described by Calkins (1970)

Most of the drums transferred to the field were nominal 55 gallon drums but a significant number were 30 gallon drums Not all were completely full Approximately three fourths of the drums were plutonium contaminated while most of the balance contained uranium Of those containing plutonium most were lathe coolant consisting of a straight chain hydrocarbon mineral oil and carbon tetrachloride in varying proportions Other liquids were involved however including hydraulic oils vacuum pump oil trichloroethylene perchloroethylene silicone oils In 1959 or possibly earlier ethanolamine was added to the oil to reduce the corrosion rate of the steel drums

An estimated 5 000 gallons of liquid (Freiberg 1970) containing 86 g (5 3 Ci) of plutonium leaked into the soil Site grading in preparation for applying an asphalt cap over the area included moving slightly contaminated soil A total of 33 drums of radioactive contaminated rocks were removed and two courses of clean fill material were placed over the site The asphalt covering was applied about two months later (Freiberg 1970) The cover is approximately 8 centimeters (cm) thick and underlain by approximately 15 cm of loose gravel and 8 cm of fill dirt

1 2 Technology Introduction

This work evaluates the use of thermally enhanced aqueous extraction processing of soils for the removal of radionuclides The concept combines the technologies of redox chelation and steam processing to meet the challenge of decontaminating 903 Pad area soils In phase I of this study emphasis was given to designing leaching schemes capable of removing Pu and Am from Rocky Flats soils The leaching schemes designed will be used to chemically enhance the steam stripping process for the mobilization and removal of radionuclide from contaminated soils This effort is part of the treatability studies for actinide-contaminated soils in OU2 s 903 Pad area but has potential applicability to other radionuclide contaminated sites at the Rocky Flats Plant

Actinides (such as Pu Am and U) tend to have a strong affinity for the minerals in most soils (Thomas 1987 and Triay et al 1991) The mechanisms that normally dominate radionuclide sorption are surface complexation (Combes et al 1992) and ion exchange (Triay and Rundberg 1989 and 1987) Since the radioactive metals are Lewis acids (i e acquire electrons to reach an inert state) complexants that act as Lewis bases (i e have electron pairs that can be shared with the metal) can be utilized to leach Pu and Am from contaminated soils

The effectiveness of complexants to remove plutonium and americium from soils depends on the chemical form of these metals in the contaminated area Pimpl and Schucttelkopf (1991) report that Pu Am and Cm in soil columns contaminated with 5 mCi of each actinide near the surface were mobilized and migrated with an irrigation solution containing 0 1M DTPA After elution of the irrigation solution less than 3 pCi/g of activity was found in the soil Lee and Marsh (1992) have reported that a significant amount of uranium can be extracted from Fernald soils utilizing citric acid and carbonate

2 0 CONCLUSIONS AND RECOMMENDATIONS

2 1 Conclusions

The results of the experiments performed indicated that using complexants alone at 20 C over 60% of the americium and 50% of the plutonium in the soil can be removed Combining complexants with a reducing or oxidizing agent yields the best results More than 70 % of the americium and 70% of the plutonium in Rocky Flats soils can be removed at 20 C by using a complexant and a reducing agent At 20 C the largest amounts of Am were removed (between 70 80%) with those leaching schemes in which dithionite was used in a pH range of 5 4 7 4 with the following complexants citric acid NTA EGTA DTPA EDTA At 20 C the largest Pu removal (i e 75%) was accomplished with NTA / dithionite at a pH of ~5 5

For approximately 40% of the leaching schemes the removal of Am from Rocky Flats soils was found to increase with increasing temperature For approximately 60% of the leaching schemes there was no practical difference in the removal of Am with increasing temperature (i e ±10% difference) In only two leaching schemes (Na_3Citrate / 3 % H_2O_2 and NTA / 3 33 eq NaOH / $Na_2S_2O_8$) was there a decrease in percent Am removal with increasing temperature The same trends were observed for Pu with the exception of the following two leaching schemes (Na_2EDTA/1 eq NaOH / 3% H_2O_2 and DTPA / 3 45 eq NaOH / 3% H_2O_2) in which the percent Pu removal decreased with an increase in temperature

At 80°C it is possible to remove over 70% of the Am in the soil and 60% of the Pu in the soil without the utilization of a redox agent The use of complexants with redox agents at 80 C removed over 80% of the Am in the soil and over 70% of the Pu from the soil At 80 C the largest amounts of Am were removed (between 80 90%) with those leaching schemes in which dithionite was used in a pH range of 5 4 7 4 with the following complexants NTA EGTA DTPA EDTA At 80 C the largest Pu removal (i e 78 %) was accomplished with NTA / dithionite at a pH of ~5 5

The matrix that was tested in Phase II of this study consisted of citric acid and a reducing agent as a function of temperature Preliminary column experiments indicated that over 70% of the alpha activity could be removed from soil columns at 20 and 75 C by using sodium citrate in conjunction with a reducing agent such as ascorbic acid or dithionite These results corroborate those found with the batch desorption experiments

2 2 Recommendations

The results obtained from Phase I of this study allow the selection of the optimal leaching schemes to chemically enhance the steam injection process for the removal of Pu and Am from Rocky Flats soils The optimal pH range for the citrate/dithionite leaching scheme for the removal of Pu and Am seems to be 5 to 6 Increase in temperature seems to enhance the removal of Pu and Am from soils using citric acid in the absence of a redox agent Testing of a leaching scheme (involving citrate and a reducing agent) has occurred using column experiments and results suggested the removal of radionuclides from soils basically stayed constant as the temperature increased in the cases where leaching schemes involved a complexing and a redox agent In Phase II of this study the actual removal of Am and Pu from the soils using column experiments and a leaching scheme involving citric acid and a reducing agent should be measured with better analytical equipment

Results from batch desorptions (phase I) and column experiments (phase II) suggest that in combination with a redox agent citrate is a suitable complexing agent to remove Pu and Am from Rocky Flats soil Removal rates for the better leaching schemes do not significantly increase with an increase in temperature and suggest that high temperatures may not be necessary during phase III of the chemically enhanced remediation technology development

3 0 TREATABILITY STUDY APPROACH

3 1 Test Objectives and Rationale

Testing will be conducted in three phases (1) selecting promising chelator redox systems to chemically enhance the steam stripping process (using fast turnaround lab scale extraction tests) (2) further refining the list of chemical enhancers to the more promising chelator redox agent systems using bench scale soil column washing tests and (3) optimizing the most promising chelator redox systems using bench scale soil column washing tests The objectives of the three-phase study are

(1) To select appropriate chelating redox agent systems and to define test matrix and work plan for bench scale tests

(2) To perform bench scale tests to evaluate plutonium and americium mobilization and removal efficiency using suitable chelating agents and redox conditions

(3) To perform bench scale tests to optimize plutonium and americium removal using suitable conditions that minimize chemical loading and modification of soils

This report will focus on the results of Phase I of this study intended to select the appropriate chelating redox agent systems to chemically enhance the steam stripping process for the mobilization and efficient removal of plutonium and americium from Rocky Flats soils

3 2 Treatment Technology Description

3 2 1 Treatment technology

Soil decontamination and washing evaluations have been conducted at RFETS since the early 1970s and results have appeared in reports in the internal and external literature R L Olsen et al (1980) described the decontamination of the 903 Pad area following cleanup of the leaking drums in 1968 They reported that radiological contamination of 2000 300 000 dpm/100 cm^2 had penetrated the 903 Pad soils to a depth of 20 cm

Hicks and Blakeslee (1981) summarized a decade of soil characterization and bench scale attrition scrubbing studies performed at RFETS on RFETS 903 Pad Area soils Plutonium in these soils occurs in both particulate (0 2 mm mean diameter associated with soil particles) and dissolved (or perhaps colloidal) forms Wet screening and radiometric characterization of the soils showed that the majority of plutonium (and americium) was associated with soils less than 2 4 mm in particle size Attrition scrubbing of soils with hot (80 C) distilled water or aqueous solutions of chemical agents (e g H_2O_2 Na_2CO_3 NaClO Na_2SiO_3) chelators (e g oxalic acid citric acid) detergents (e g Oakite NST) and surfactants (e g sodium dioctyl sulfosuccinate) showed varying decontaminating effectiveness Oxalic acid (0 1M) sodium hexametaphosphate (10%) and 10% detergent solutions were among the more effective decontaminating systems for the 2 4-4 0 mm soil fraction removing 95 98% of the Pu and Am However residual contamination levels still exceeded 60 dpm/g (27 pCi/g) following the scrubbing process

Hicks and Blakeslee (1981) also reported soil washing tests on soils from five DOE sites including RFETS Three aqueous solutions (1) an aqueous pH 12 5 (2) 2% HNO_3 0 2% HF 2% pine oil and 5% hexametaphosphate (3) 2 N HCl were evaluated for decontaminating RFETS Hanford Mound INEL and LANL soils Variability in the effectiveness of the three phase scrubbing process was noted for soils from the different DOE sites Effectiveness of the solutions also varied with the soil size fraction tested

Pettis and Kallas (1988) conducted bench scale testing and reported that simple room temperature wet screening of 903 Pad soils was capable of decontaminating the size fraction greater than 4 mm (approximately 60 wt% of the total) to <5 dpm/g (2 3 pCi/g) Pu and Am The size fraction greater than 2 4 mm (approximately 65 wt% of the total) was decontaminated to less than 12 dpm/g (5 5 pCi/g) Pu and 6 dpm/g (2 7 pCi/g) Am by wet attrition scrubbing The remaining (soil fraction less than 2 4 mm) was treated by attrition scrubbing ultrasonic scrubbing oxidation calcination desliming flotation and heavy liquid density separation Although somewhat guarded because of results for selected size fractions they concluded that attrition or vibratory scrubbing and either mineral jig or acid leaching of this fraction would be effective for a decontamination goal of <30 dpm/g (14 pCi/g)

Steam Stripping

Of the various remediation approaches noted above under the thermal category of remediation technologies one innovative technology is dynamic underground stripping—which is an adaptation of steam injection and electrical heating Steam injection accelerates removal of the NAPL contaminants and is combined with vacuum extraction to perform accelerated removal of volatile contaminants such as underground hydrocarbon spills electrical heating accelerates the process Steam injection technology has been demonstrated for remediating NAPLs and VOCs in subsurface soils and clay layers Aines and Newmark (1992) and Buettner et al (1992) have successfully tested this technology in combination with electrical heating on a bench scale and small field scale for the removal of NAPLs/VOCs in soils or clay layers EPA s Vendor Information Service for Innovative Treatment Technologies (VISITT) reports that steam stripping technology is being commercialized by Praxis Environmental Services (San Francisco CA) (Stewart 1992)

Redox Chemistry

Mobilization of contaminants can be obtained as a result of physical or chemical action Chemically induced mobilization occurs either by chemical action on the contaminant directly (e g plutonium) or its support substrate (i e soil) In the case of plutonium contaminated soils the mobilization of soil bound species depends to a large extent on the physical and chemical properties of both the plutonium and the soil

Plutonium generally exists in multiple oxidation states III IV V and VI and in the natural environment (associated with soils) plutonium is normally found in either the IV state or to a lesser extent the VI state The oxidation state of the plutonium is a determining factor in its solubility — III V and VI states being more soluble than the IV state whereas americium normally occurs only in the III state Redox (or chemical *red*uction/*ox*idation) behavior of plutonium features highly in its propensity for dissolution and modification of its oxidation state is an important tool in changing solubility Cleveland (1971) has described conditions for both reduction and oxidation of Pu(IV) using a variety of chemical reagents We evaluated simple reducing and oxidizing agents and conditions to accelerate dissolution/mobilization of the plutonium

Chelation Chemistry

Mobilization of metallic contaminants is greatly enhanced by formation of strong attachments with chemical binding agents via chelation Chelation chemistry and chelators (or chelates) are often used in association with metals to accelerate dissolution and/or stabilize/maintain solubility by diminishing the tendency to readsorb or precipitate Once dissolved the soluble species (in this case radionuclide-chelator complexes) are stabilized by their association with the chelate which in turn enhances their continued mobility A variety of complexing or chelating agents including EDTA are commonly used for this purpose Chelating agents can also enhance dissolution by tightly binding the radionuclide and preventing readsorption or precipitation

Chemically Enhanced Steam Stripping

The remediation technology under development combines steam injection redox and chelation for the leaching/washing of radionuclides from soils Steam injection/extraction technology enhanced by redox and chelation chemistry were used to mobilize and flush away radionuclides contaminating RFETS soils Conceptually the process is very similar in design to steam injection systems involving injection and downgradient extraction wells but with novel modifications to the injection system to provide chemical addition upstream of the soil washing regime

The technology is refined to minimize chemical injection with the goal of conducting *in situ* evaluation or *ex situ* treatment with the eventual return of the treated soils to the site The benefits of this approach include (1) reducing the volume of contaminated soils (2) avoiding extremely harsh conditions therefore improving the potential for soil post treatment soil viability and/or replacement (3) possible tailoring to contaminant and soil conditions and (4) combining with stream stripping of VOCs

3 3 Experimental Design and Procedures

3 3 1 Batch Desorption Experiments

Batch desorption experiments involved sieving the soil samples to particle size of less than 53 mm adding a leaching solution containing a complexant and/or a redox agent mixing the two phases separating the phases and determining the amount of plutonium and americium in each phase These desorption experiments were conducted at both 20 C and 80 C All desorption experiments were performed in duplicate The soil used for these experiments had a very small particle size (less than 53 mm) the reason for this choice is that the Pu and Am content in this size fraction is larger than in the larger sizes and harder to mobilize The complexants tested as a function of pH were citric acid NTA EDTA EGTA DTPA and carbonate The reducing agents were ascorbic acid and dithionite the oxidizing agents were hydrogen peroxide the hypochlorite ion and the persulfate ion Combinations of the complexants with reducing or oxidizing agents were also tested The preparation of the leaching solutions used is given in Appendix A The chemical structure of the complexants and redox agents is given in Appendix B and C respectively

3 3 2 Column Experiments

Four leaching solutions (sodium citrate sodium citrate with ascorbic acid sodium citrate with dithionite and sodium chloride solutions) were eluted at 20 C through columns containing RFETS soil Air dried RFETS soil samples were sieved through a 2 mm sieve prior to use in the column experiments The soil column experiments were performed in duplicate The amount of radionuclide leached from the RFETS soil was determined by total alpha counting The columns used contain 350 g of air dried 2 mm sieved RFETS soil At least ten column volumes were eluted to determine the amount of radionuclides leached from the soil

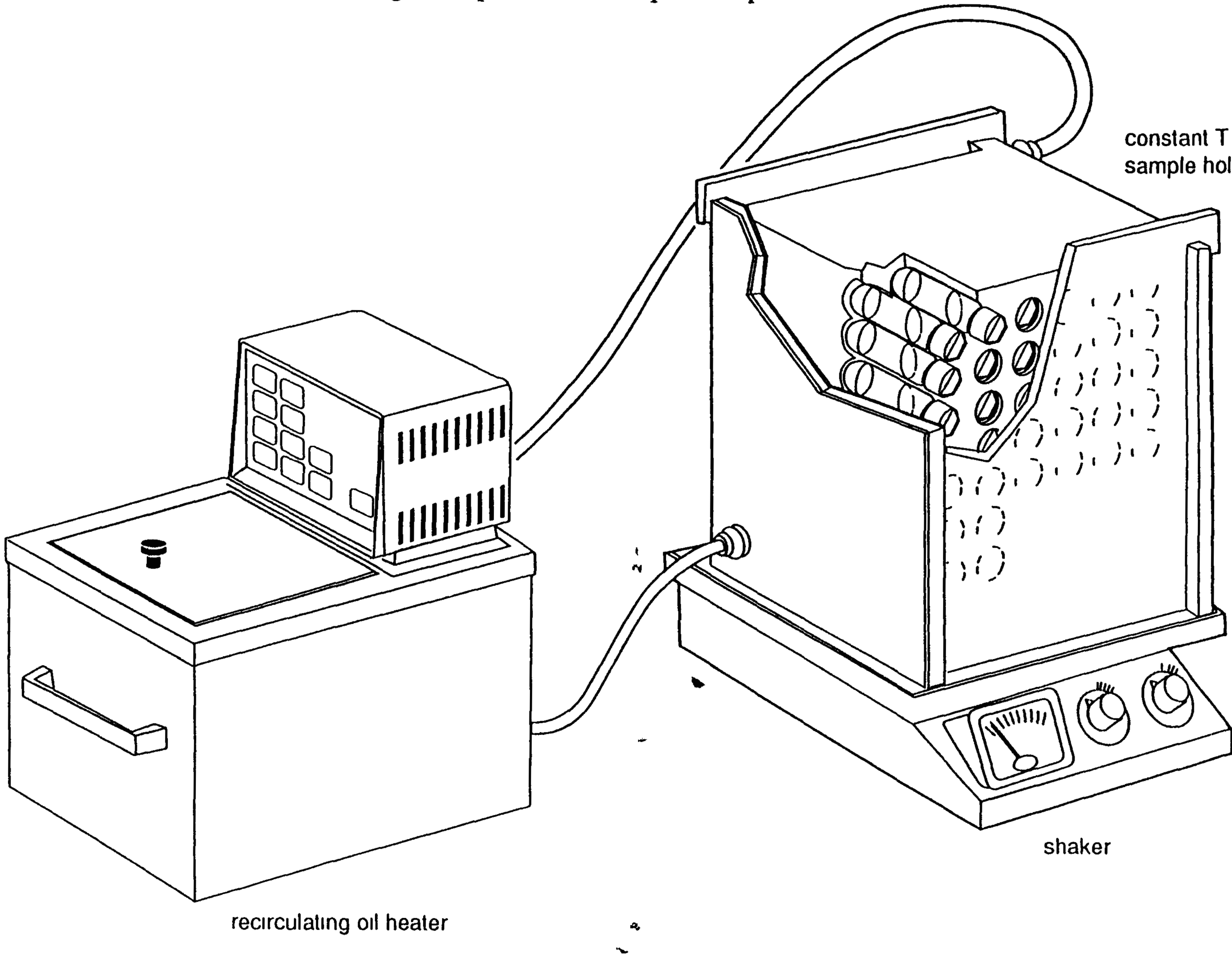
3 4 Detail Experimental Procedures

3 4 1 Batch Desorption Experiments

The desorption experiments at 20 C consisted of 1) weighing 2 5 g of the RFETS sieved soil (with a particle size of <53 mm) into an Oak Ridge centrifuge tube 2) adding 20 ml of a freshly prepared leaching solution to the soil in the tube and capping the tube tightly 3) placing the tube in an orbital shaker for 24 hours 4) centrifuging the sample for one hour at 12 000 rpm (28 000 g) 5) decanting the liquid from the solid into a fresh centrifuge tube 6) centrifuging the decanted liquid for one hour at 12 000 rpm 7) pipetting 10 mL of the centrifuged liquid into a third centrifuge tube for a final two hour centrifugation cycle at 12 000 rpm 8) analyzing 5 mL of the centrifuged liquid for ^{239}Pu and ^{241}Am content (using alpha and gamma spectrometry respectively) 9) weighing the wet solid sample (left from step 5) 10) drying the wet solid under a heat lamp for 48 hours then weighing again to obtain the weight of the residual extractant liquid left behind from the decanting procedure and 11) analyzing the dried soil samples for ^{239}Pu and ^{241}Am content using gamma spectrometry

For the 80 C desorption experiments extra care was taken during transferring and centrifuging so that the temperature would be maintained as close to 80 C as possible To that end the pipette tips OR tubes and the centrifuge rotor used were preheated to ~100°C The desorption experiments at 80 C consisted of 1) weighing 2 5 g of the RFETS sieved soil (with a particle size of <53 mm) into an Oak Ridge centrifuge tube 2) adding 20 ml of a freshly prepared leaching solution to the soil in the tube and capping the tube tightly 3) placing the tube in a specially designed silicone oil surrounded shaker (see Figure 1) whose temperature had been equilibrated to 80 C for 24 hours 4) quickly transferring the tube containing the equilibrated solution into an ~100 C preheated centrifuge rotor and centrifuging for 10 minutes at 20 000 rpm 5) quickly and carefully decanting the liquid into a preheated fresh tube 6) placing the tube containing the decanted liquid into a preheated centrifuge rotor and centrifuging for 10 minutes at 20 000 rpm 7) quickly pipetting 16 ml of the centrifuged liquid into a fresh preheated tube 8) placing the tube containing the pipetted liquid into a preheated centrifuge rotor and centrifuging for 10 minutes at 20 000 rpm 9) pipetting 5 mL of centrifuged solution into a tared vial containing 1 mL of fresh extractant (to try to prevent precipitation) 10) analyzing the diluted centrifuged liquid for ^{239}Pu and ^{241}Am content (using alpha and gamma spectrometry respectively) 11) weighing the wet solid sample (left from step 5) 12) drying the wet solid under a heat lamp for 48 hours then weighing again to obtain the weight of the residual extractant liquid left behind from the decanting procedure and 13) analyzing the dried soil samples for ^{239}Pu and ^{241}Am content using gamma spectrometry

Figure 1 Apparatus Used for High Temperature Desorption Experiments



Gamma spectrometry analysis was performed using a 180 cc HPGe detector with a well (1 cm in diameter and 5 cm deep) into which a Plexiglas capsule with the sample is placed This geometry permits capturing about 90% of ^{241}Am gammas and about 60% of the ^{239}Pu gammas All dry solid samples had a very intense ^{241}Am 59 5 KeV peak and a much weaker but unmistakably identifiable ^{239}Pu 129 5 KeV peak The reason why the Pu peak is weaker than the Am one is that only 0 006 alpha decays of ^{239}Pu are accompanied by a 129 5 KeV photon whereas about half of ^{241}Am decays give a 59 5 KeV photon Each dry solid sample was measured for about 4 hours The ^{239}Pu concentration in the solid samples was measured by gamma spectrometry with 20 30% accuracy in most cases the ^{241}Am concentration was measured with much better accuracy in both solid and liquid samples

3 4 2 Column Experiments

Column experiments were conducted using selected leaching solutions Kontes Chromaflex™ Chromatography glass columns with safety water jackets were used There are two layers in each column The inner column is made from glass and the contaminated Rocky Flats soil was packed into it The outer column is made from acrylic plastic and used for temperature control A peristaltic pump was used to pump leaching solution into the soil column

The size of the inner column is 4 8 cm in diameter and 15 cm in length About 350 g of soil were loaded into each column about 20 g of washed sand were placed on the top and bottom of the column to improve the drainage of the washing solution from the soil column The sand had been washed twice with 5 % HCl solution and once with 30% H_2O_2 solution then rinsed 3 times with distilled water and air dried The flow rate in the columns was about 1ml/min After leaching the columns for four days with the desired solution the columns were washed with 0 1 M sodium chloride The soil in the columns was then removed and air dried The dried soil sample was counted for radioactivity by alpha counting A scintillation alpha counter (Model SAC 4 Eberline Instrument Company Santa Fe NM) was used Soil samples were counted at least four times for 20 minutes The percent removal based on total alpha activity is reported in 4 1 2

The soil pH was measured on a slurry that was 1 part soil and 1 part water

3 5 Sampling and Data Management

Representativeness in the amount of plutonium in the soil to be analyzed is a difficult goal to obtain It is extremely time consuming to dissolve large amounts of soil prior to alpha spectrometry in order to determine the exact amount of plutonium in each phase Therefore the soils received were air dried and sieved prior to utilization in these experiments Assuming that the plutonium is associated with a particular size fraction in the RFETS soils choosing a narrow size fraction (that contains a large amount of plutonium) alleviates the problem of representativeness in sampling and analysis

All soils received from RFETS were stored and handled according to the guidelines of LANL procedures for Storage and Handling of Solid Samples

4 0 RESULTS AND DISCUSSION

4 1 Data Analysis and Interpretation

4 1 1 Batch Desorption Experiment

The details and raw data used for the results presented in this section can be found in Appendix D The results of the batch desorption experiments performed at 20 C and 80 C are given in Table 1 for complexants and reducing or oxidizing agents

Table 1 Americium and Plutonium Removal from Rocky Flats Soils at 20 C and 80 C for Distilled Water and Reducing and Oxidizing Agents

Extractant	eq NaOH	eq H_2SO_4	pH at 20°	pH at 80°	/ Am Removal at 20°	/ Am Removal at 80	/ Pu Removal at 20	/ Pu Removal at 80
DI Water	0	0	80	79	0	1	1	1
01 M Ascorbic Acid	0	0	37	39	1	10	3	10
01 M Ascorbic Acid	1	0	6.2	66	27	45	22	
01 M $Na_2S_2O_4$	0	0	53	40	0	1	1	0
3/ H_2O_2	0	0	49	47	0	0	7	1
01 M $Na_2S_2O_8$	0	0	27	19	2	66	1	25
5/ NaOCl	0	0	78	70	2	6	9	15
1/ NaOCl	0	040	39	49	5	5	7	5

Inspection of Table 1 indicates that plain deionized water removes a negligible amount of Am and Pu from the soil The utilization of oxidizing and reducing agents by themselves (without a complexant) did not accomplish significant removal of actinides from the soil at 20 or 80 C These results were expected and the leaching experiments with the redox agents (without a complexant) were performed for completeness

The differences observed between the Am and Pu removal from soil are probably due to the initial oxidation states of the actinides in the solid phase Americium is likely to exist in the III oxidation state whereas Pu is likely in the IV oxidation state Table 2 indicates that complexants (in the absence of oxidizing or reducing agents) are more successful for the removal of Am than Pu The results obtained from Table 2 are graphically displayed in Appendix E (% Removal as a Function of pH) Appendix F (% Removal as a Function of Redox Agent) and Appendix G (% Removal as a Function of Lixiviant with all Redox Agents Used)

Table 2 Americium and Plutonium Removal from Rocky Flats Soils at 20 C and 80 C for Complexing Agents as a Function of pH and Added Reducing and Oxidizing Agents

Complexing Agent	eq NaOH	eq H_2SO_4	Redox Agent	pH at 20°	pH at 80°	/ Am Removal at 20	/ Am Removal at 80°	/ Pu Removal at 20	/ Pu Removal at 80
0 1 M Citric Acid	0	0		2.2	2 4	29	36	28	63
	1	0		3.5	3.5	51	52	56	57*
	2	0		4 6	4 9	60	71	36	58
(Na_3Citrate)	3	0		8 3	8 7	29	42◊	20	35◊
0 1 M Na_3Citrate	0	0	Ascorbic Acid	5 4	5 4	69	73	56	51
	1	0	Ascorbic Acid	6 8	7.2	52	61	38	40
	0	0	Na2S2O4	5 9	6 4	71	67	62	57
	0 53	0	$Na_2S_2O_4$	5 6	6 7	63	61	63	53
0 3 M Na_3Citrate / 0 01 M $NaHCO_3$	0	0	0 5 M $Na_2S_2O_4$	6 1	5 8	77	70	52	
0 1 M Na_3Citrate	0	0	3 / H_2O_2	8 4	8 3	54	42	48*	40
	0	0	$Na_2S_2O_8$	6 9	5 1	54	77	45	57
	0	0 76	1 / NaOCl	8 6	9.2	33	41	39	37
0 1 M EGTA	2	0		4 8	5 0	58	69		42
	3	0		8 7	8 7	56	67	23	31
	3 5	0		9.2	9.2	59	64	26	27
	4	0		9 9	9 8	53	64	17	28
	3 27	0	Ascorbic Acid	5 9	6.2	61	76	37	53
	2 89	0	$Na_2S_2O_4$	6 0	6 4	75	81	62	70*
	2 30	0	3 / H_2O_2	4 0	4 3	57	68	27	46
	2.30	0	$Na_2S_2O_8$	3.2	2 9	53	72	26	48
	0	2.38	1 / NaOCl	8 1	7.2	27	67	12	43

Table 2 Continued

Complexing Agent	eq NaOH	eq H_2SO_4	Redox Agent	pH at 20°	pH at 80°	/ Am Removal at 20	' Am Removal at 80	' Pu Removal at 20°	Pu Removal at 80
0 1 M Na_2EDTA	0	0		4 6	5 6	67	75	30	57
	1	0		7 7	8.3	60	72	20	42
	2	0		10 5	10 3	55	64	18	33
	2	0	Ascorbic Acid	6 0	7 4	67	77	60*	59*
	1	0	$Na_2S_2O_4$	6.2	6.5	73	83	55	60
	1	0	3/ H_2O_2	5 9	7.5	69	62	54	28
	1	0	$Na_2S_2O_8$	4 8	6 3	61	56	32	39
	0 83	0	1/ NaOCl	7 9	8 4	57	68	30	29
0 1 M DTPA	2	0		3 9	3 8	65	79	41	
	3	0		5.2	6 7	61	75	44	48
	3.37	0		7 0	7 4	60	74	26	52
	4	0		8 7	8 9	60	66◊	24	64◊
	5	0		10 3	10.3	60	66	24	38
	4 50	0	Ascorbic Acid	6 7	6 9	66	76	50	
	4 32	0	$Na_2S_2O_4$	7 4	6.5	71	86	56	68
	3 45	0	3/ H_2O_2	6 6	7 4	65	70	50	38
	3.50	0	$Na_2S_2O_8$	4 0	5 1	64	77	42	
	4 01	0	1/ NaOCl	7 4	8 1	60	70	31	54

Table 2 Continued

Complexing Agent	eq NaOH	eq H_2SO_4	Redox Agent	pH at 20°	pH at 80°	/ Am Removal at 20°	/ Am Removal at 80	/ Pu Removal at 20	' Pu Removal at 80
0 05 M NTA	1	0		3.2	5 7	59	69	53	51
0 08 M NTA	1.5	0		3.5	5 4	63	75	47	56
0 1 M NTA	2	0		5 4	6 3	62	75	44	44
	2.5	0		7.5	7 4	59	71	28	58
	3	0		10 1	9 7	59	62	29	22
	3 33	0	Ascorbic Acid	5 7	7 0	63	75	55	61
	2 68	0	$Na_2S_2O_4$	5 4	6.3	75	80	75	78
	2.22	0	3/ H_2O_2	8 8	7 7	58	65	26	43
	2.18	0	$Na_2S_2O_8$	4 1	2 9	60	32	40*	
	2.20	0	1/ NaOCl	7 9	8 1	50	62	42	36
2 M Na2CO3	0	0		11 0	10 8	26	20	17	18
0 1 M Na2CO3	0	0		10 4	10 1	1	3	2	2
0 1 M NaHCO3	0	0		8 7	8 6	0	1	0	1
	0.22	0	Ascorbic Acid	7.2	7 1	25	42	13	28
	0 12	0	$Na_2S_2O_4$	6 6	7 1	0	1	1	2
	0 015	0	3/ H_2O_2	8 9	9 1	1	0	1	1
	0	0	$Na_2S_2O_8$	8.3	2.2	0	54	2	
	0	0 37	1/ NaOCl	8.3	8.5	2	3	4	8

All duplicate trials of the leaching schemes found in table 2 were within 10% of each other except those marked with an ◊ or those left blank An * indicates that the duplicate trials were within 11 15% and ◊ indicates that data for only one trial was available Spaces left blank indicate that the results obtained on duplicate runs were >15% and need to be repeated

In general the complexants are most effective removing Pu and Am from the soils in the pH range from 3 5 to 5 Basic solutions reduce the removal of Pu from the soils significantly One possible explanation for this observation is the formation of hydroxides at large pH values followed by precipitation of the actinides rather than solubilization and removal from the soil into the solution phase

The results from Table 2 indicate that using complexants alone at 20 C over 60% of the americium and 50% of the plutonium in the soil can be removed Combining complexants with a reducing or oxidizing agent yields the best results More than 70% of the americium and 70% of the plutonium in Rocky Flats soils can be removed at 20 C by using a complexant and a reducing agent At 20 C the largest amounts of Am were removed (between 70 80%) with those leaching schemes in which dithionite was used in a pH range of 5 4 7 4 with the following complexants citric acid NTA EGTA DTPA EDTA At 20 C the largest Pu removal (i e 75%) was accomplished with NTA / dithionite at a pH of ~5 5

For approximately 40% of the leaching schemes the removal of Am from Rocky Flats soils was found to increase with increasing temperature For approximately 60% of the leaching schemes there was no practical difference in the removal of Am with increasing temperature (i e ±10% difference) In only two leaching schemes (Na_3Citrate / 3% H_2O_2 and NTA / 3 33 eq NaOH / $Na_2S_2O_8$) was there a decrease in percent Am removal with increasing temperature The same trends were observed for Pu with the exception of the following two leaching schemes (Na_2EDTA/1 eq NaOH / 3% H_2O_2 and DTPA / 3 45 eq NaOH / 3% H_2O_2) in which the percent Pu removal decreased with an increase in temperature

At 80°C it is possible to remove over 70% of the Am in the soil and 60% of the Pu in the soil without the utilization of a redox agent The use of complexants with redox agents at 80 C removed over 80% of the Am in the soil and over 70% of the Pu from the soil At 80 C the largest amounts of Am were removed (between 80-90%) with those leaching schemes in which dithionite was used in a pH range of 5 4 7 4 with the following complexants NTA EGTA DTPA EDTA At 80°C the largest Pu removal (i e 78%) was accomplished with NTA / dithionite at a pH of ~5 5

Given the formation of actinide-carbonate complexes in solution one would expect sodium carbonate to strongly complex the actinides in the soil inspection of Table 2 indicates that this is not the case at the concentrations of sodium carbonate used (from 0 1 to 2 M) Even with 2 M sodium carbonate only 26% and 17% of the Am and Pu in the soils respectively were removed

The soils utilized for the leaching experiments were sieved to a size of less than 53 mm The initial concentration of the soils (< 53 mm in particle size) used for the 20 °C experiments (originally from Drum ASI52594-01 Plot 28) was 800 pCi/g and 5 100 pCi/g for ^{241}Am and ^{239}Pu respectively The initial concentration of the soils used for the 80 °C experiments (originally from Drum ASI52594 03 Plot 28) was 600 pCi/g and 4,500 pCi/g for ^{241}Am and ^{239}Pu respectively Since both the soil and the solution phases were counted durıng the leachıng experıments ıt was possıble to calculate the total amount of Am and Pu ın the alıquots of soıls taken for the desorptıon experıments The amount of ^{241}Am ın the soıls from the 20°C experıments was 800 ± 46 pCı/g the amount of ^{239}Pu ın the soıls was 5 200 ± 870 pCı/g The amount of ^{241}Am ın the soıls from the 80°C experıments was 700 ± 35 pCı/g the amount of ^{239}Pu ın the soıls was 4 500 ± 700 pCı/g The uncertaınty assocıated wıth these measurement not only represents countıng errors but the heterogeneıty of the actınıde concentratıons ın the soıl

4 1 2 Column Experıments

Table 3 summarızes the results of column experıments performed wıth Rocky Flats soıls The removal of alpha actıvıty from the soıl was attempted usıng varıous leachıng schemes All the column experıments were performed at room temperature and 75 °C

Table 3 Results of Column Studıes

Leachıng Solutıon	/ Alpha Actıvıty Removed @ 20 C	/ Alpha Actıvıty Removed @ 75 C	Startıng Soıl pH	Fınal Soıl pH
0 1 M Sodıum Chlorıde	1/	1/	6 8	6 4
0 1 M Sodıum Cıtrate	22/	38/	6 8	8.3
0 1 M Sodıum Cıtrate with 0 1 M Sodıum Dıthıonıte	80/	75/	6 8	6 7
0 1 M Sodıum Cıtrate wıth 0 1 M Ascorbıc Acıd	70/	78/	6 8	6 4

Column results were consistent with batch desorption data Results suggest that little or no radioactivity was removed from the soil by leaching with 0 1 M sodium chloride solution at 20 C and 75 C About 20% of the radioactivity could be washed out by using 0 1 M sodium citrate An increase in temperature would enhance the removal of radionuclides from soils using a citrate solution in the absence of a redox agent However only about 40% of radionuclides were removed by using sodium citrate alone as a leaching solution About 70 80% of the radioactivity could be removed from the soil column by using sodium citrate in conjunction with a reducing agent such as ascorbic acid or the dithionite ion The removal rate of radionuclides from soils stayed basically constant (within the error of the analytical procedure used) as the temperature increased from 20 C and 75 C in the cases where the leaching solution contained a reducing agent It should be noted that the radioactivity removal rates were measured using total alpha counting and there may be about 15 % potential experimental error resulting from the utilization of this method

4 2 Quality Assurance And Control

Work performed in this project is governed by the Work Plan for Chemically Enhanced Stream Stripping of Radionuclides in RFETS Soils Quality Assurance Requirements and applicable Standard Operating Procedures which have been approved by the Los Alamos National Laboratory Quality Assurance Organization All batch desorption and column experiments were performed in duplicate The data obtained were stored in spreadsheets using Microsoft Excel Hard copies of the spreadsheets which include the data obtained and any procedural deviations are signed by the technician performing the work and the principal investigator Electronic copies of the spreadsheets are stored on a hard disk drive which is backed up on to an optical disk on a weekly basis All the procedures used follow quality assurance guidelines

REFERENCES

R D Aines R L Newmark Rapid Removal of Underground Hydrocarbon Spills *Energy & Technology Review* UCRL 52000 92 7 (July 1992) p 1 7

H M Buettner W D Daily A L Ramirez Enhancing Cyclic Steam Injection and Vapor Extraction of Volatile Organic Compounds in Soils with Electrical Heating UCRL ID 109424 Lawrence Livermore National Laboratory Livermore CA March 30 1992

Calkins K W Memorandum to L M Joshel Dow Chemical Company Rocky Flats Division August 19 1970

Cleveland J W The Chemistry of Plutonium Gordon and Breach Science Publishers New York 1970

Combes J M C J Chisholm Brause G E Brown Jr G A Parks S D Conradson P G Eller I R Triay D E Hobart and A Meijer X ray Absorption Spectroscopy Study of Neptunium (V) Sorbed at the a FeOOH/Water Interface *Environmental Science and Technology* 26 376-382 (1992)

K J Freiberg Memorandum to E A Putzier Dow Chemical Company Rocky Flats Division April 14 1970

J E Hicks J J Blakeslee Soil Decontamination Process Development Closeout Report AR 05-15 20 1 AL Rockwell International Rocky Flats Plant Golden CO September 1981

Lee S Y and J D Marsh Jr Characterization of Uranium Contaminated Soils from DOE Fernald Environmental Management Project Site Results of Phase I Characterization Oak Ridge National Laboratory Report ORNL/TM 11980 (January/1992)

R L Olsen J A Hayden C E Alford R L Kochen J R Stevens Soil Decontamination at Rocky Flats *Decontamination and Decommissioning of Nuclear Facilities* M M Osterhout Ed Plenum Publishing Corp 1980 p 161 172

S A Pettis A J Kallas Summary of Previous Soil Decontamination Studies Performed at Rocky Flats Plant PSD88 0027 Rockwell International Rocky Flats Plant Golden CO June 1988

Pimpl M and H Schucttelkopf Decontamination of Soils by Irrigation with Solutions Containing Complexing Agents preprint

L D Stewart K S Udell Thermally Enhanced Recovery In Situ Praxis Environmental Services (San Francisco CA) from EPA s Vendor Information System for Innovative Treatment Technologies (VISITT) database Version 2 0 1992

Thomas K Summary of Sorption Measurements Performed with Yucca Mountain Nevada Tuff Samples and Water from Well J 13 Los Alamos National Laboratory Report LA 10960 MS (December/1987)

Triay I R A J Mitchell and M A Ott Radionuclide Migration as a Function of Mineralogy in Proceedings of the Second Annual International Conference on High-Level Radioactive Waste Management April 28 May 3 1991 Las Vegas NV Vol 1 pp 494-498

Triay I R and R S Rundberg Deconvolution of Multivalent Cation Exchange Isotherms *Journal of Physical Chemistry* 93 5617 5623 (1989)

Triay I R and R S Rundberg Application of Deconvolution to the Analysis of Univalent Ion Exchange Isotherms in Zeolites X and Y *Zeolites* 9 ,217 23 (1989)

Triay I R and R S Rundberg Determination of Selectivity Coefficient Distributions by Deconvolution of Ion Exchange Isotherms *Journal of Physical Chemistry* 91 5269 74 (1987)

Appendix A Preparation of Leaching Solutions

0 100 M Ascorbic Acid 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Ascorbic Acid / 1 eq NaOH 1 76 g of ascorbic acid (FW = 176 1) and 10 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M $Na_2S_2O_4$ 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

3% H_2O_2 Taken directly from reagent bottle

0 100 M $Na_2S_2O_8$ 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

5% NaOCl Taken directly from clorox bleach bottle

1% NaOCl / 0 40 eq H_2SO_4 16 7 mL of 6% NaOCl and 4 0 mL of 1 00 N H_2SO_4 were dissolved in enough water to produce 100 mL of solution

0 100 M Citric Acid 2 10 g of Citric acid monohydrate (FW = 210 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Citric Acid / 1 eq NaOH 2 10 g of Citric acid monohydrate (FW = 210 0) and 10 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Citric Acid / 2 eq NaOH 2 10 g of Citric acid monohydrate (FW = 210 0) and 20 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate 2 94 g of sodium citrate dihydrate (FW = 294 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate / 0 100 M Ascorbic Acid 2 94 g of sodium citrate dihydrate (FW = 294 1) and 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate / 1 eq NaOH / 0 100 M Ascorbic Acid 2 94 g of sodium citrate dihydrate (FW = 294 1) 10 0 mL of 1 00 M NaOH and 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate / 0 100 M $Na_2S_2O_4$ 2 94 g of sodium citrate dihydrate (FW = 294 1) and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate / 0 53 eq NaOH / 0 100 M $Na_2S_2O_4$ 2 94 g of sodium citrate dihydrate (FW = 294 1) 5 3 mL of 1 00 M NaOH and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

0 267 M Sodium Citrate / 0 011 M $NaHCO_3$ / 0 480 M $Na_2S_2O_4$ 7 85 g of sodium citrate dihydrate (FW = 294 1) 0 0932 g of sodium bicarbonate (FW = 84 0) and 8 34 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate / 3 % H_2O_2 2 94 g of sodium citrate dihydrate (FW = 294 1) were dissolved in enough 3 % hydrogen peroxide to produce 100 mL of solution

0 100 M Sodium Citrate / 0 100 M $Na_2S_2O_8$ 2 94 g of sodium citrate dihydrate (FW = 294 1) and 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Sodium Citrate / 0 76 eq H_2SO_4 / 1% NaOCl 2 94 g of sodium citrate dihydrate (FW = 294 1) and 7 6 mL of 1 00 N H_2SO_4 were dissolved in enough 1% NaOCl to produce 100 mL of solution

0 100 M EGTA / 2 eq NaOH 3 80 g of Ethyleneglycol-O O bis(2 aminoethyl) N N,N N tetraacetic acid (FW = 380 4) and 20 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce100 mL of solution

0 100 M EGTA / 3 eq NaOH 3 80 g of Ethyleneglycol-O O bis(2 aminoethyl) N N,N N tetraacetic acid (FW = 380 4) and 30 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce100 mL of solution

0 100 M EGTA / 3 5 eq NaOH 3 80 g of Ethyleneglycol-O O bis(2 aminoethyl)-N N N',N tetraacetic acid (FW = 380 4) and 35 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce100 mL of solution

0 100 M EGTA / 4 eq NaOH 3 80 g of Ethyleneglycol-O O bis(2 aminoethyl) N N N N tetraacetic acid (FW = 380 4) and 40 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce100 mL of solution

0 100 M EGTA / 3 27 eq NaOH / 0 100 M Ascorbic Acid 3 80 g of Ethyleneglycol-O O bis(2 aminoethyl) N N N N' tetraacetic acid (FW = 380 4) 32 7 mL of 1 00 M NaOH and 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M EGTA / 2 89 eq NaOH / 0 100 M $Na_2S_2O_4$ 3 80 g of Ethyleneglycol-O O bis(2 aminoethyl) N N,N N tetraacetic acid (FW = 380 4) 2 89 mL of 1 00 M NaOH and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M EGTA / 2 30 eq NaOH / 3 % H_2O_2 3 80 g of Ethyleneglycol-O O bis(2 aminoethyl) N N,N N' tetraacetic acid (FW = 380 4) and 23 0 mL of 1 00 M NaOH were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution

0 100 M EGTA / 2 30 eq NaOH / 0 100 M $Na_2S_2O_8$ 3 80 g of Ethyleneglycol-O O bis(2 aminoethvl) N N N N tetraacetic acid (FW = 380 4) 23 0 mL of 1 00 M NaOH and 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M EGTA / 2 38 eq NaOH / 1/ NaOCl 3 80 g of Ethyleneglycol O O bis(2 aminoethyl) N N N N tetraacetic acid (FW = 380 4) and 23 8 mL of 1 00 M NaOH were dissolved in enough 1 / NaOCl to produce 100 mL of solution

0 100 M Na_2EDTA 3 72 g of the Disodium salt of ethylenediamine tetraacetic acid dihydrate (FW = 372 2) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Na_2EDTA / 1 eq NaOH 3 72 g of the Disodium salt of ethylenediamine tetraacetic acid dihydrate (FW = 372 2) and 10 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Na_2EDTA / 2 eq NaOH 3 72 g of the Disodium salt of ethylenediamine tetraacetic acid dihydrate (FW = 372 2) and 20 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Na_2EDTA / 2 eq NaOH / 0 100 M Ascorbic Acid 3 72 g of the disodium salt of ethylenediamine tetraacetic acid (FW = 372 2) 20 0 mL of 1 00 M NaOH and 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Na_2EDTA / 1 eq NaOH / 0 100 M $Na_2S_2O_4$ 3 72 g of the disodium salt of ethylenediamine tetraacetic acid (FW = 372 2) 10 0 mL of 1 00 M NaOH and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution.

0 100 M Na_2EDTA / 1 eq NaOH / 3% H_2O_2 3 72 g of the disodium salt of ethylenediamine tetraacetic acid (FW = 372 2) and 10 0 mL of 1 00 M NaOH were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution

0 100 M Na_2EDTA / 1 eq NaOH / 0 100 M $Na_2S_2O_8$ 3 72 g of the disodium salt of ethylenediamine tetraacetic acid (FW = 372 2) 10 0 mL of 1 00 M NaOH and 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Na_2EDTA / 0 83 eq NaOH / 1% NaOCl 3 72 g of the disodium salt of ethylenediamine tetraacetic acid (FW = 372 2) and 8 3 mL of 1 00 M NaOH were dissolved in enough 1% NaOCl to produce 100 mL of solution

0 100 M DTPA / 2 eq NaOH 3 93 g of Diethylenetriaminepentaacetic acid (FW = 393 3) and 20 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 3 eq NaOH 3 93 g of Diethylenetriaminepentaacetic acid (FW = 393 3) and 30 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 3 37 eq NaOH 3 93 g of Diethylenetriaminepentaacetic acid (FW = 393 3) and 33 7 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 4 eq NaOH 3 93 g of Diethylenetriaminepentaacetic acid (FW = 393 3) and 40 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 5 eq NaOH 3 93 g of Diethylenetriaminepentaacetic acid (FW = 393 3) and 50 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 4 50 eq NaOH / 0 100 M Ascorbic Acid 3 93 g of Diethylenetriaminepentaacetic acid (FW = 393 3) 45 0 mL of 1 00 M NaOH and 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 4 32 eq NaOH / 0 100 M $Na_2S_2O_4$ 3 93 g of Diethylene-triaminepentaacetic acid (FW = 393 3) 43 2 mL of 1 00 M NaOH and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 3 45 eq NaOH / 3% H_2O_2 3 93 g of Diethylenetriamine pentaacetic acid (FW = 393 3) and 34 5 mL of 1 00 M NaOH were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution

0 100 M DTPA / 3 50 eq NaOH / 0 100 M $Na_2S_2O_8$ 3 93 g of Diethylene triaminepentaacetic acid (FW = 393 3) 35 0 mL of 1 00 M NaOH and 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M DTPA / 4 01 eq NaOH / 1% NaOCl 3 93 g of Diethylenetriamine pentaacetic acid (FW = 393 3) and 40 1 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 050 M NTA / 1 eq NaOH 0 955 g of Nitrilotriacetic acid dihydrate (FW = 191 1) and 10 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 080 M NTA / 1 5 eq NaOH 1 53 g of Nitrilotriacetic acid dihydrate (FW = 191 1) and 15 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M NTA / 2 eq NaOH 1 91 g of Nitrilotriacetic acid dihydrate (FW = 191 1) and 20 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M NTA / 2 5 eq NaOH 1 91 g of Nitrilotriacetic acid dihydrate (FW = 191 1) and 25 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M NTA / 3 eq NaOH 1 91 g of Nitrilotriacetic acid dihydrate (FW = 191 1) and 30 0 mL of 1 00 M NaOH were dissolved in enough distilled water to produce 100 mL of solution

0 100 M NTA / 3 33 eq NaOH / 0 100 M Ascorbic Acid 1 91 g of Nitrilotriacetic acid (FW = 191 1) 33 3 mL of 1 00 M NaOH and 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M NTA / 2 68 eq NaOH / 0 100 M $Na_2S_2O_4$ 1 91 g of Nitrilotriacetic acid (FW = 191 1) 26 8 mL of 1 00 M NaOH and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M NTA / 2 22 eq NaOH / 3% H_2O_2 1 91 g of Nitrilotriacetic acid (FW = 191 1) and 22 2 mL of 1 00 M NaOH were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution

0 100 M NTA / 2 18 eq NaOH / 0 100 M $Na_2S_2O_8$ 1 91 g of Nitrilotriacetic acid (FW = 191 1) and 21 8 mL of 1 00 M NaOH and 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M NTA / 2 20 eq NaOH / 1% NaOCl 1 91 g of Nitrilotriacetic acid (FW = 191 1) and 22 0 mL of 1 00 M NaOH were dissolved in enough 1% NaOCl to produce 100 mL of solution

2 00 M Na_2CO_3 21 2 g of sodium carbonate (FW = 106 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M Na_2CO_3 1 06 g of sodium carbonate (FW = 106 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M $NaHCO_3$ 0 840 g of sodium carbonate (FW = 84 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M $NaHCO_3$ / 0 22 eq NaOH / 0 100 M Ascorbic Acid 0 840 g of Sodium bicarbonate (FW = 84 01) 2 2 mL of 1 00 M NaOH and 1 76 g of ascorbic acid (FW = 176 1) were dissolved in enough distilled water to produce 100 mL of solution

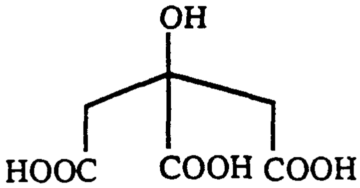
0 100 M $NaHCO_3$ / 0 12 eq NaOH / 0 100 M $Na_2S_2O_4$ 0 840 g of Sodium bicarbonate (FW = 84 01) 1 2 mL of 1 00 M NaOH and 1 74 g of sodium dithionite (FW = 174 1) were dissolved in enough distilled water to produce 100 mL of solution.

0 100 M $NaHCO_3$ / 0 015 eq NaOH / 3% H_2O_2 0 840 g of Sodium bicarbonate (FW = 84 01) and 0 15 mL of 1 00 M NaOH were dissolved in enough 3% hydrogen peroxide to produce 100 mL of solution

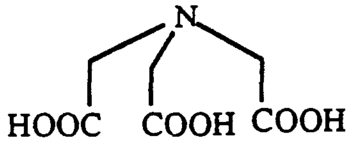
0 100 M $NaHCO_3$ / 0 100 M $Na_2S_2O_8$ 0 840 g of Sodium bicarbonate (FW = 84 01) and 2 38 g of sodium persulfate (FW = 238 0) were dissolved in enough distilled water to produce 100 mL of solution

0 100 M $NaHCO_3$ / 0 37 eq H_2SO_4 */ 1% NaOCl* 0 840 g of Sodium bicarbonate (FW = 84 01) and 3 7 mL of 1 00 N H_2SO_4 were dissolved in enough 1% NaOCl to produce 100 mL of solution

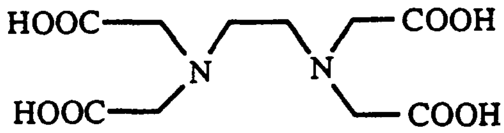
Appendix B Complexants



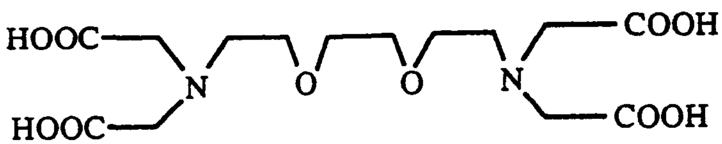
Citric Acid



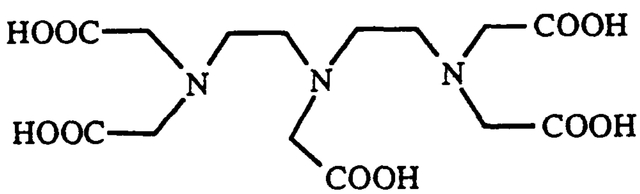
NTA
Nitrilotriacetic Acid



EDTA
Ethylenediamine tetracetic acid



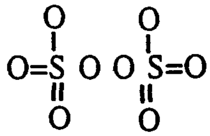
EGTA
Ethyleneglycol O O bis(aminoethyl) N N,N',N' tetraacetic acid



DTPA
Diethylenetriaminepentaacetic acid

Appendix C Oxidizing and Reducing Agents

Oxidizing Agents



persulfate ion

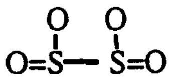
H O O H

hydrogen peroxide

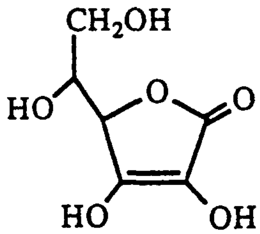
NaOCl

bleach (sodium hypochlorite)

Reducing Agents



dithionite ion



ascorbic acid

Appendix D

Data for Batch Desorptions at 20° and 80°

[NOTE For the column in which the Average Percent Removal of Radionuclide is given almost all of the duplicates were within 10% of each other unless otherwise marked An indicates that duplicate trials were within 11 15 % a ◊ indicates that data for only one trial was available and a duplicate needs to be run and a † indicates that the results obtained on duplicate runs were greater than 15% and the experiment needs to be repeated]

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RTP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
Citric Acid	147	#11	2 50	19 66	1 8	322	2 2	301	1 72	2 11	6 10
	148	#11	2 51	20 06			2 2	298	1 82	2 50	5 97
Citric Acid / 1 eq NaOH	149	#11	2 50	20 10	3 2	343	3 5	269	1 63	2 44	6 13
	150	#11	2 55	20 19			3 5	270	1 75	2 60	5 95
Citric Acid / 2 eq NaOH	151	#11	2 51	20 20	4 4	269	4 6	239	1 63	2 48	5 93
	152	#11	2 54	20 20			4 6	243	1 56	2 48	5 92
Sodium Citrate	153	#11	2 50	20 25	8 0	234	8 3	104	1 58	2 44	5 97
	154	#11	2 55	20 22			8 2	103	1 69	2 41	6 00
EGTA / 2 eq NaOH	155	#11	2 50	20 15	6 0	318	4 8	136	1 47	2 45	5 98
	156	#11	2 51	20 17			4 8	137	1 63	2 47	5 94
EGTA / 3 eq NaOH	157	#11	2 51	20 37	8 9	167	8 7	5	1 55	2 47	6 01
	158	#11	2 51	20 33			8 7	5	1 59	2 46	6 01
EGTA / 3 5 eq NaOH	159	#11	2 50	20 33	9 2	71	9 2	19	1 64	2 48	6 00
	160	#11	2 50	20 31			9 1	18	1 58	2 47	6 00
EGTA / 4 eq NaOH	161	#11	2 50	19 88	9 9	44	9 9	41	1 59	2 47	5 96
	162	#11	2 52	19 77			9 9	-40	1 54	2 51	6 07

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Citric Acid	147	640 ± 38	29 ± 3	3 039 ± 683	138 ± 14	27	29	848	27	28	4 029
	148	587 ± 34	31 ± 3	3 664 ± 719	183 ± 21	31		812	29		4 994
Citric Acid / 1 eq NaOH	149	445 ± 26	52 ± 4	1 860 ± 684	281 ± 24	50	51	829	57	56	3 936
	150	456 ± 27	58 ± 4	2 374 ± 767	320 ± 27	52		875	54		4 688
Citric Acid / 2 eq NaOH	151	392 ± 23	66 ± 5	4 108 ± 695	230 ± 19	60	60	881	32	36	5 811
	152	390 ± 23	67 ± 5	3 110 ± 580	251 ± 18	60		884	40		4 951
Sodium Citrate	153	642 ± 38	32 ± 3	3 052 ± 653	111 ± 14	29	29	881	23	20	3 882
	154	690 ± 40	34 ± 2	3 791 ± 551	89 ± 10	29		937	16		4 438
EGTA / 2 eq NaOH	155	387 ± 22	59 ± 4	3 440 ± 712	257 ± 24	57	58	827	39	29 †	5 357
	156	385 ± 22	59 ± 4	4 682 ± 709	140 ± 18	58		821	20		5 717
EGTA / 3 eq NaOH	157	373 ± 22	58 ± 3	3 239 ± 690	103 ± 14	58	56	809	21	23	4 012
	158	429 ± 25	58 ± 3	4 272 ± 757	170 ± 21	55		862	25		5 542
EGTA / 3 5 eq NaOH	159	352 ± 21	61 ± 3	3 137 ± 695	140 ± 14	61	59	808	27	26	4 183
	160	400 ± 23	59 ± 4	3 596 ± 634	137 ± 18	57		843	24		4 624
EGTA / 4 eq NaOH	161	427 ± 25	55 ± 3	5 239 ± 697	130 ± 14	53	53	829	17	17	6 190
	162	397 ± 23	54 ± 3	4 046 ± 673	110 ± 18	54		788	18		4 843

Appendix D Data for Batch Experiment at 20 C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFI (20°C)	Gram of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
Na2EDTA	163	#12	2 50	20 29	4 9	268	4 6	224	1 68	2 46	6 02
	164	#12	2 50	20 26			4 6	228	1 73	2 42	6 00
Na2EDTA / 1 eq NaOH	165	#12	2 52	20 35	8 1	171	7 7	87	1 85	2 44	6 13
	166	#12	2 54	20 32			7 7	81	1 92	2 49	6 15
Na2EDTA / 2 eq NaOH	167	#12	2 49	20 41	11 0	57	10 5	4	2 35	2 43	6 11
	168	#12	2 51	20 36			10 4	2	2 25	2 44	5 98
DTPA / 2 eq NaOH	169	#12	2 49	20 41	3 3	340	3 9	192	1 78	2 47	5 92
	170	#12	2 53	20 26			3 9	190	1 87	2 46	6 39
DTPA / 3 eq NaOH	171	#12	2 53	20 30	4 9	286	5 2	106	1 93	2 47	6 13
	172	#12	2 54	20 40			5 2	102	1 62	2 50	6 04
DTPA / 4 eq NaOH	173	#12	2 50	20 48	8 9	123	8 7	-4	1 97	2 46	5 88
	174	#12	2 50	20 43			8 7	-6	1 89	2 44	5 95
DTPA / 5 eq NaOH	175	#12	2 55	20 47	10 4	34	10 3	-43	1 81	2 51	5 93
	176	#12	2 50	20 50			10 3	-43	1 78	2 49	6 05
DI Water	177	#12	2 52	19 94	5 7	70	7 7	105	2 19	2 46	5 76
	178	#12	2 50	19 93			8 3	103	2 31	2 45	5 99

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Na2EDTA	163	329 ± 19	71 ± 4	4 011 ± 653	205 ± 19	67	67	858	30	30	5 538
	164	320 ± 18	69 ± 4	3 953 ± 667	205 ± 18	67		831	30		5 472
Na2EDTA / 1 eq NaOH	165	358 ± 21	62 ± 4	3 488 ± 665	132 ± 14	62	60	814	24	20	4 458
	166	391 ± 23	59 ± 3	5 511 ± 897	127 ± 18	58		819	16		6 433
Na2EDTA / 2 eq NaOH	167	444 ± 26	59 ± 3	4 555 ± 872	127 ± 14	55	55	871	19	18	5 475
	168	428 ± 25	55 ± 3	3 872 ± 732	102 ± 13	54		825	18		4 607
DTPA / 2 eq NaOH	169	335 ± 20	69 ± 4	4 473 ± 916	330 ± 27	66	65	852	39	41	6 945
	170	342 ± 20	65 ± 4	4 038 ± 770	357 ± 29	64		815	43		6 636
DTPA / 3 eq NaOH	171	381 ± 22	68 ± 4	2 916 ± 520	300 ± 27	62	61	875	47	44	5 097
	172	396 ± 23	66 ± 4	3 448 ± 731	275 ± 27	60		883	40		5 477
DTPA / 4 eq NaOH	173	382 ± 22	62 ± 3	4 185 ± 722	140 ± 16	60	60	841	22	24	5 222
	174	391 ± 23	62 ± 4	3 997 ± 811	164 ± 16	60		851	26		5 215
DTPA / 5 eq NaOH	175	399 ± 23	62 ± 4	4 083 ± 684	137 ± 14	58	60	853	22	24	5 087
	176	348 ± 20	58 ± 3	3 935 ± 811	160 ± 16	61		782	26		5 131
DI Water	177	760 ± 45	*0 1* ±	5 153 ± 747	11 ± 10	0	0	761	2	1	5 230
	178	767 ± 45	*0 1* ±	5 194 ± 718	*5* ±	0		768	1		5 229

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RJT (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
2 M Sodium Carbonate	179	#13	2 53	23 49	11 4	33	11 0	9	1 62	2 81	6 78
	180	#13	2 50	23.50			11 0	0	1 61	2 81	7 28
Sodium Carbonate	181	#13	2 51	20 11	11 2	176	10 4	30	1 76	2 46	6 36
	182	#13	2 58	20 09			10 4	26	1 79	2 53	6 29
Sodium Bicarbonate	183	#13	2 48	19 94	8 8	396	8 7	99	1 78	2 43	6 27
	184	#13	2 57	19 93			8 7	120	1 94	2 51	6 27
0 05 M NTA / 1 eq NaOH	185	#13	2 55	20 06	2 6	386	3 2	295	1 73	2 46	6 31
	186	#13	2 53	21 30			3 1	323	1 72	2 44	6 28
0 08 M NTA / 1 5 eq NaOH	187	#13	2 48	20 54	2 8	386	3 5	290	1 90	2 41	6 03
	188	#13	2 53	20 04			3 5	291	1 99	2 46	6 01
NTA / 2 eq NaOH	189	#13	2 56	20 09	3 6	344	5 4	224	2 22	2 51	6 06
	190	#13	2 50	20 13			5 4	215	1 95	2 44	6 06
NTA / 2 5 eq NaOH	191	#13	2 56	20 19	8 6	178	7 5	141	2 46	2 52	6 04
	192	#13	2 54	20 20			7 5	133	2 43	2 51	6 05
NTA / 3 eq NaOH	193	#13	2 47	20 18	10 3	61	10 1	3	2 04	2 41	5 88
	194	#13	2 48	20 16			10 0	36	2 35	2 43	6 07

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
2 M Sodium Carbonate	179	502 ± 29	19 ± 1	4 153 ± 611	68 ± 14	26	26	662	13	17	4 741
	180	497 ± 29	18 ± 1	2 709 ± 552	78 ± 5	26		658	22		3 392
Sodium Carbonate	181	863 ± 51	2 ± 0	7 056 ± 933	16 ± 4	1	1	874	2	2	7 173
	182	838 ± 51	1 ± 0	6 426 ± 1093	14 ± 1	1		847	2		6 525
Sodium Bicarbonate	183	809 ± 48	*0 1* ±	4 611 ± 704	*5* ±	0	0	810	1	0	4 648
	184	805 ± 48	*0 1* ±	5 986 ± 814	0 4 ± 0 1	0		806	0		5 989
0 05 M NTA / 1 eq NaOH	185	359 ± 21	62 ± 4	2 730 ± 518	424 ± 51	61	59	805	58	53	5 782
	186	396 ± 23	59 ± 4	3 000 ± 529	305 ± 40	58		853	48		5 364
0 08 M NTA / 1 5 eq NaOH	187	377 ± 26	67 ± 4	3 125 ± 540	337 ± 40	63	63	880	49	47	5 655
	188	371 ± 22	67 ± 4	3 432 ± 513	329 ± 40	63		849	45		5 778
NTA / 2 eq NaOH	189	382 ± 22	66 ± 4	3 159 ± 497	332 ± 40	61	62	842	48	44	5 474
	190	378 ± 22	68 ± 5	3 943 ± 534	316 ± 43	63		872	41		6 239
NTA / 2 5 eq NaOH	191	389 ± 23	61 ± 4	3 990 ± 520	237 ± 37	59	59	811	33	28	5 631
	192	413 ± 24	63 ± 4	4 338 ± 551	151 ± 32	59		854	22		5 395
NTA / 3 eq NaOH	193	391 ± 23	57 ± 4	3 558 ± 631	203 ± 37	57	59	809	33	29	5 047
	194	407 ± 24	63 ± 4	4 695 ± 624	183 ± 35	60		860	25		6 012

Appendix D Data for Batch Experiments at 20 C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RIT (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
Ascorbic Acid	195	#14	2.51	19.98	2.7	137	3.7	5	1.81	2.41	6.35
	196	#14	2.51	20.02			3.7	2	1.71	2.45	6.31
Dithionite	197	#14	2.52	20.08	3.2	45	5.2	67	2.01	2.43	6.43
	198	#14	2.52	20.09			5.3	47	1.99	2.44	6.34
Hydrogen Peroxide	199	#14	2.50	19.81	4.2	369	4.9	175	1.97	2.23	6.12
	200	#14	2.52	19.53			4.8	170	2.04	2.27	5.94
NaOCl	201	#14	2.53	21.59	10.9	751	7.9	156	1.46	2.52	6.76
	202	#14	2.52	21.49			7.6	179	1.58	2.49	6.82
Persulfate	203	#14	2.51	20.24	4.3	61	2.7	589	1.95	2.48	6.07
	204	#14	2.53	20.27			2.7	606	1.83	2.49	6.06
Na3Citrate / Ascorbic Acid	205	#14	2.49	19.62	5.1	31	5.4	103	2.47	2.51	6.11
	206	#14	2.51	19.79			5.4	103	2.49	2.51	6.09
Na3Citrate / Dithionite	207	#14	2.51	20.43	5.2	353	5.9	110	2.00	2.48	6.10
	208	#14	2.51	20.55			5.9	107	1.87	2.50	6.14
Na3Citrate / Hydrogen Peroxide	209	#14	2.51	20.02	7.5	235	8.0	32	2.48	2.27	6.07
	210	#14	2.50	19.82			8.7	18	2.45	2.20	6.08

Appendix D Data for Batch Experiments at 20 C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Ascorbic Acid	195	810 ± 48	2 ± 0	3 315 ± 570	18 ± 8	2	1	823	4	3	3 445
	196	831 ± 49	1 ± 0	4 391 ± 694	10 ± 2	1		835	2		4 464
Dithionite	197	818 ± 48	0.3 ±	5,559 ± 720	8 ± 8	0	0	820	1	1	5 616
	198	780 ± 46	0 3 ±	4,536 ± 696	2 2 ± 0 5	0		782	0		4 552
Hydrogen Peroxide	199	799 ± 47	0 3 ±	5 128 ± 782	35 ± 16	0	0	801	5	7	5 378
	200	813 ± 48	0.3 ±	3 945 ± 640	51 ± 10	0		815	9		4 299
NaOCl	201	819 ± 48	2 5 ± 0 3	4 660 ± 732	32 ± 20	3	2	839	6	9	4 915
	202	828 ± 49	1 9 ± 0 2	3 296 ± 615	54 ± 5	2		843	12		3 723
Persulfate	203	735 ± 43	1 8 ± 0 4	5 015 ± 726	5 ± 5	2	2	748	1	1	5 051
	204	783 ± 48	1 5 ± 0 2	4,275 ± 505	2 4 ± 0 8	2		794	0		4 293
Na3Citrate / Ascorbic Acid	205	310 ± 18	68 ± 5	2 975 ± 512	394 ± 48	69	69	778	55	56	5 689
	206	313 ± 18	72 ± 5	2 290 ± 430	332 ± 35	70		809	57		4 576
Na3Citrate / Dithionite	207	299 ± 17	70 ± 5	2 671 ± 508	440 ± 45	70	71	814	61	62	5 907
	208	291 ± 17	71 ± 5	2 427 ± 511	437 ± 64	71		819	63		5 676
Na3Citrate / Hydrogen Peroxide	209	417 ± 24	55 ± 3	2 340 ± 713	314 ± 40	55	54	802	55	48	4 539
	210	441 ± 26	57 ± 4	3 842 ± 744	299 ± 35	54		838	40		5 923

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
Ascorbic Acid / 1 eq NaOH	211	#15	2 49	20 10	5 6	92	6 1	145	1 91	2 46	6 41
	212	#15	2 48	20 12			6 2	151	2 01	2 44	6 23
Na3Cit / 1 eq NaOH / Ascorbic Acid	213	#15	2 54	20 48	6 8	153	6 8	163	2 43	2 55	6 47
	214	#15	2 51	20 50			6 7	158	2 51	2 53	6 41
Na3Citrate / Persulfate	215	#15	2 47	20 52	7 7	338	7 1	363	2 20	2 48	5 98
	216	#15	2 51	20 54			6 6	360	2 19	2 49	6 56
0 3 M Na3Cit / 0 01 M NaHCO3 / 0 5 M Dithionite	217	#15	2 48	21 98	5 9	500	6 1	149	1 69	2 61	7 19
	218	#15	2 54	22 06			6 1	162	1 55	2 64	6 91
Na2EDTA / 2 eq NaOH / Ascorbic Acid	219	#15	2 52	20 48	7 5	212	6 0	167	2 04	2 52	6 10
	220	#15	2 53	20 56			5 9	170	2 22	2 55	6 05
Na2EDTA / 1 eq NaOH / Dithionite	221	#15	2 50	20 65	5 5	-440	6 2	0	2 05	2 52	6 20
	222	#15	2 54	20 74			6 2	0	2 01	2 57	6 37
Na2EDTA / 1 eq NaOH / Hydrogen Peroxide	223	#15	2 51	20 51	7 2	258	5 8	200	4 74	2 52	5 07
	224	#15	2 49	20 53			5 9	198	5 38	2 52	5 87
Na2EDTA / 1 eq NaOH / Na2S2O8	225	#15	2 50	20 61	4 1	325	4 8	379	2 29	2 57	5 88
	226	#15	2 52	20 67			4 8	382	2 76	2 64	6 12

Appendix D Data for Batch Experiments at 20 C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Ascorbic Acid / 1 eq NaOH	211	597 ± 35	27 ± 2	3 314 ± 521	127 ± 24	27	27	794	24	22	4 241
	212	587 ± 34	25 ± 2	4 103 ± 597	116 ± 21	26		770	19		4 952
Na3Cit / 1 eq NaOH / Ascorbic Acid	213	413 ± 24	49 ± 3	4 331 ± 497	278 ± 40	52	52	761	36	38	6 306
	214	402 ± 23	48 ± 3	4 473 ± 434	343 ± 40	53		746	40		6 934
Na3Citrate / Persulfate	215	423 ± 25	52 ± 3	3 646 ± 606	318 ± 40	53	54	808	44	45	6 002
	216	404 ± 24	54 ± 4	3 077 ± 527	289 ± 40	55		799	46		5 192
0 3 M Na3Cit / 0 01 M NaHCO3 / 0 5 M Dithionite	217	224 ± 13	70 ± 5	2 481 ± 576	256 ± 43	78	77	797	50	52	4 576
	218	243 ± 14	74 ± 5	1 832 ± 521	235 ± 29	76		840	55		3 728
Na2EDTA / 2 eq NaOH / Ascorbic Acid	219	352 ± 20	70 ± 5	1 850 ± 601	386 ± 43	66	67	863	67	60	4 670
	220	325 ± 19	72 ± 5	3 096 ± 641	394 ± 54	69		848	54		5 958
Na2EDTA / 1 eq NaOH / Dithionite	221	371 ± 22	75 ± 6	2 683 ± 484	378 ± 43	67	73 *	928	57	55	5 492
	222	249 ± 14	88 ± 6	2 822 ± 437	354 ± 43	80		898	53		5 434
Na2EDTA / 1 eq NaOH / Hydrogen Peroxide	223	370 ± 21	73 ± 5	2 714 ± 509	340 ± 43	72	69	828	57	54	4 849
	224	424 ± 25	68 ± 4	3 533 ± 564	340 ± 48	67		839	50		5 606
Na2EDTA / 1 eq NaOH / Na2S2O8	225	348 ± 20	60 ± 4	4 628 ± 452	299 ± 33	63	61	788	36	32	6 823
	226	423 ± 25	62 ± 4	4 749 ± 469	211 ± 46	59		864	28		6 249

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RIT (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
DTPA / 3 37 eq NaOH	227	#16	2 53	20 11	8 0	132	7 0	32	1 66	2 46	6 12
	228	#16	2 52	20 07			7 0	35	1 68	2 46	6 08
DTPA / 4 50 eq NaOH / Ascorbic Acid	229	#16	2 48	20 48	8 1	245	6 7	178	1 59	2 42	6 09
	230	#16	2 52	20 63			6 7	173	1 62	2 50	6 12
DTPA / 4 32 eq NaOH / Na2S2O4	231	#16	2 48	20 80	7 8	-460	7 4	43	1 67	2 51	6 20
	232	#16	2 50	20 80			7 4	46	1 63	2 49	6 21
DTPA / 3 5 eq NaOH / Hydrogen Peroxide	233	#16	2 49	20 58	7 5	260	6 5	289	3 03	2 51	6 14
	234	#16	2 50	20 71			6 6	291	3 55	2 50	6 15
DTPA /3 50 eq NaOH / Na2S2O8	235	#16	2 50	16 94	3 8	119	4 0	171	1 69	2 53	5 65
	236	#16	2 54	16 84			4 0	148	1 61	2 54	6 40
NTA / 3 33 eq NaOH / Ascorbic Acid	237	#16	2 53	20 12	8 2	275	5 9	128	1 89	2 53	5 88
	238	#16	2 51	20 21			5 4	61	1 98	2 50	6 38
NTA / 2 68 eq NaOH / Na2S2O4	239	#16	2 49	20 51	6 4	392	5 4	74	1 52	2 44	6 41
	240	#16	2 49	20 64			5 3	81	1 54	2 46	6 33
NTA / 2 22 eq NaOH / H2O2	241	#16	2 47	20 40	8 1	226	8 8	51	2 18	2 38	6 23
	242	#16	2 52	20 39			8 8	47	2 02	2 40	5 20

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu-239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
DTPA / 3 37 eq NaOH	227	407 ± 24	64 ± 4	5 148 ± 508	210 ± 24	58	60	874	25	26	6 681
	228	377 ± 22	66 ± 4	5 102 ± 484	229 ± 27	61		859	27		6 773
DTPA / 4 50 eq NaOH / Ascorbic Acid	229	325 ± 19	69 ± 5	3 096 ± 524	399 ± 43	67	66	850	54	50	6 130
	230	340 ± 20	68 ± 5	3 523 ± 515	346 ± 49	65		853	46		6 133
DTPA / 4 32 eq NaOH / Na2S2O4	231	299 ± 17	72 ± 5	2 387 ± 477	262 ± 37	71	71	854	50	56 *	4 405
	232	290 ± 17	70 ± 5	2 602 ± 490	465 ± 49	70		827	63		6 168
DTPA / 3 5 eq NaOH / Hydrogen Peroxide	233	364 ± 21	66 ± 4	2 879 ± 435	272 ± 32	66	65	829	47	50	4 796
	234	382 ± 22	64 ± 4	2 160 ± 439	256 ± 48	65		821	54		3 917
DTPA /3 50 eq NaOH / Na2S2O8	235	340 ± 20	73 ± 5	3 788 ± 541	316 ± 32	63	64	785	37	42	5 713
	236	339 ± 20	78 ± 5	2 659 ± 494	318 ± 46	64		808	46		4 569
NTA / 3 33 eq NaOH / Ascorbic Acid	237	366 ± 21	65 ± 4	3 090 ± 543	441 ± 67	62	63	835	56	55	6 269
	238	349 ± 20	65 ± 4	2 875 ± 490	365 ± 62	64		821	53		5 527
NTA / 2 68 eq NaOH / Na2S2O4	239	285 ± 16	77 ± 5	1 272 ± 451	398 ± 48	73	75	871	76	75	4 303
	240	229 ± 13	78 ± 5	1 769 ± 451	489 ± 73	78		828	73		5 523
NTA / 2 22 eq NaOH / H2O2	241	481 ± 28	64 ± 4	5 557 ± 707	202 ± 32	56	58	954	24	26	7 051
	242	378 ± 22	61 ± 4	4 449 ± 648	213 ± 32	60		822	29		5 999

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
NTA / 2 18 eq NaOH / Na2S2O8	243	#17	2 51	19 65	6 8	448	4 1	431	1 56	5 01	5 32
	244	#17	2 51	19 70			4 1	440	1 53	5 17	6 25
NaHCO3/0 22 eq NaOH/Ascorbic Acid	245	#17	2 50	19 88	8 1	230	7 2	222	1 77	5 05	5 43
	246	#17	2 51	19 98			7 2	227	1 69	4 84	6 23
NaHCO3/0 12 eq NaOH/Na2S2O4	247	#17	2 52	20 17	6 1	90	6 6	61	1 84	4 45	6 27
	248	#17	2 51	20 12			6 6	63	1 76	4 48	6 21
NaHCO3 / Na2S2O8	249	#17	2 51	19 52	9 1	332	8 3	315	1 65	5 21	6 20
	250	#17	2 51	20 19			8 3	322	1 74	4 49	6 21
EGTA / 3 27 eq NaOH / Ascorbic Acid	251	#17	2 51	20 49	8 0	223	5 9	147	1 88	4 39	6 18
	252	#17	2 50	20 61			5 9	148	1 95	4 05	6 13
EGTA / 2 89 eq NaOH / Na2S2O4	253	#17	2 51	20 30	6 0	114	6 0	1	1 62	4 43	6 22
	254	#17	2 51	20 39			6 0	-4	1 71	4 46	6 24
EGTA / 2 30 eq NaOH/Na2S2O8	255	#17	2 50	20 65	3 3	302	3 3	351	1 89	4 04	6 19
	256	#17	2 51	20 66			3 1	358	1 77	4 05	6 19
Na3Cit / 0 53 eq NaOH / Na2S2O4	257	#17	2 50	20 48	5 7	22	5 6	7	1 72	4 18	6 23
	258	#17	2 51	20 54			5 6	0	1 64	4 32	5 65

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
NTA / 2 18 eq NaOH / Na2S2O8	243	364 ± 21	66 ± 4	2,303 ± 528	246 ± 33	62	60	840	47	40 *	4 078
	244	373 ± 22	62 ± 4	3 257 ± 581	195 ± 30	59		822	33		4 670
NaHCO3/0 22 eq NaOH/Ascorbic Acid	245	655 ± 39	26 ± 2	3 802 ± 562	72 ± 13	25	25	843	13	13	4 324
	246	604 ± 35	24 ± 2	4 873 ± 631	81 ± 13	25		779	12		5 465
NaHCO3/0 12 eq NaOH/Na2S2O4	247	803 ± 47	0 3 ±	4 879 ± 519	2 3 ± 0 4	0	0	805	0	1	4 896
	248	783 ± 46	0 3 ±	4 262 ± 501	3 8 ± 0 8	0		785	1		4 290
NaHCO3 / Na2S2O8	249	761 ± 45	0 3 ±	3 705 ± 406	8 ± 1	0	0	763	2	2	3 761
	250	778 ± 46	0 3 ±	3 714 ± 417	8 ± 1	0		780	2		3 773
EGTA / 3 27 eq NaOH / Ascorbic Acid	251	374 ± 22	58 ± 4	3 774 ± 509	316 ± 33	59	61	804	42	37	6 119
	252	356 ± 21	65 ± 4	4 721 ± 544	262 ± 24	64		841	32		6 675
EGTA / 2 89 eq NaOH / Na2S2O4	253	243 ± 14	71 ± 5	2 652 ± 546	385 ± 45	74	75	772	57	62	5 523
	254	256 ± 15	74 ± 5	2 332 ± 454	492 ± 54	75		808	67		6 002
EGTA / 2 30 eq NaOH/Na2S2O8	255	393 ± 23	48 ± 3	3 682 ± 567	157 ± 17	53	53	753	27	26	4 860
	256	397 ± 23	52 ± 4	2 791 ± 540	112 ± 24	54		789	25		3 635
Na3Cit / 0 53 eq NaOH / Na2S2O4	257	345 ± 20	65 ± 5	2 172 ± 484	405 ± 43	64	63	832	64	63	5 209
	258	353 ± 21	64 ± 4	2 393 ± 415	424 ± 38	63		835	62		5 587

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
NaHCO3 / 0 015 eq NaOH / 3% H2O2	259	#18	1 32	9 90	8 3	177	9 0	114	1 04	13 35	5 17
	260	#18	1 31	9 97			8 7	113	1 09	13 54	4 88
EGTA / 2 30 eq NaOH / 3% H2O2	261	#18	1 30	10 40	6 6	284	4 0	205	1 04	12 97	5 09
	262	#18	1 30	10 36			4 0	196	1 10	13 15	4 87
1% NaOCl / 0 40 eq H2SO4	263	#18	1 32	10 24	6 5	975	3 9	252	0 96	13 02	5 13
	264	#18	1 31	10 24			3 9	259	0 99	13 13	5 71
Na3Citrate / 0 76 eq H2SO4 / 1% NaOCl	265	#18	1 31	10 57	8 6	465	8 6	37	1 18	1 34	5 15
	266	#18	1 30	10 43			8 6	51	1 17	1 34	5 25
Na2EDTA / 0 83 eq NaOH / 1% NaOCl	267	#18	1 31	10 55	7 4	110	7 9	45	1 21	12 84	5 88
	268	#18	1 30	10 48			7 9	48	1 19	13 01	5 00
DTPA / 4 01 eq NaOH / 1% NaOCl	269	#18	1 30	10 67	7 6	88	7 4	13	1 29	12 86	5 31
	270	#18	1 30	10 55			7 4	12	1 26	13 01	5 16
NTA / 2 20 eq NaOH / 1% NaOCl	271	#18	1 30	10 54	7 3	104	7 8	43	1 19	12 91	5 42
	272	#18	1 31	10 44			7 9	40	1 19	12 90	5 00
NaHCO3 / 0 37 eq H2SO4 / 1% NaOCl	273	#18	1 30	10 44	7 6	888	8 3	128	1 06	12 84	5 07
	274	#18	1 30	10 38			8 3	121	1 09	12 93	5 07

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
NaHCO3 / 0 015 eq NaOH / 3% H2O2	259	976 ± 57	1 ± 0	5 859 ± 741	11 ± 11	1	1	983	1	1	5 933
	260	970 ± 57	2 ± 0	6 311 ± 692	11 ± 11	1		980	1		6 385
EGTA / 2 30 eq NaOH / 3% H2O2	261	442 ± 26	61 ± 4	4 757 ± 572	222 ± 30	55	57	882	28	27	6 359
	262	404 ± 24	61 ± 4	5 319 ± 556	227 ± 38	58		839	26		6 937
1% NaOCl / 0 40 eq H2SO4	263	850 ± 50	7 ± 1	4 713 ± 602	54 ± 16	6	5	900	8	7	5 093
	264	1068 ± 63	5 ± 1	5 264 ± 614	43 ± 16	3		1102	6		5 567
Na3Citrate / 0 76 eq H2SO4 / 1% NaOCl	265	610 ± 36	35 ± 3	2 563 ± 411	140 ± 22	33	33	862	32	31	3 858
	266	572 ± 34	33 ± 2	2,537 ± 392	132 ± 22	33		807	30		3 918
Na2EDTA / 0 83 eq NaOH / 1% NaOCl	267	409 ± 24	57 ± 4	3 855 ± 458	205 ± 21	56	57	816	31	30	5 320
	268	439 ± 26	63 ± 4	3 647 ± 466	173 ± 40	57		891	29		4 888
DTPA / 4 01 eq NaOH / 1% NaOCl	269	403 ± 23	62 ± 4	4 105 ± 541	250 ± 25	60	60	851	35	31	5 913
	270	389 ± 23	62 ± 4	4 843 ± 480	208 ± 40	60		832	27		6 330
NTA / 2 20 eq NaOH / 1% NaOCl	271	419 ± 26	51 ± 3	4 389 ± 545	348 ± 43	53	50	786	41	42	6 895
	272	437 ± 26	46 ± 3	3 759 ± 539	319 ± 57	48		761	42		6 006
NaHCO3 / 0 37 eq H2SO4 / 1% NaOCl	273	891 ± 52	2 ± 0	5 047 ± 527	18 ± 3	2	2	903	3	4	5 177
	274	903 ± 53	3 ± 1	5 238 ± 481	30 ± 5	3		924	4		5 453

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RIP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of liquid sent for analysis
EGTA / 2 38 eq H2SO4 / 1% NaOCl	275	#18	1 34	10 28	7 9	90	8 1	186	1 27	13 09	5 03
	276	#18	1 31	10 12			8 0	186	1 31	13 40	5 04

Appendix D Data for Batch Experiments at 20°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
EGTA / 2 38 eq H2SO4 / 1% NaOCl	275	729 ± 43	32 ± 2	6 437 ± 758	103 ± 11	26	27	944	11	12	7 130
	276	646 ± 38	32 ± 2	5 647 ± 707	111 ± 35	29		862	13		6 396

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
Citric Acid	279	#19	2 53	19 24	1 8	322	2 3	215	1 87	2 41	5 88	4 91
	280	#19	2 54	19 32			2 4	214	1 44	2 42	5 92	4 95
Citric Acid / 1 eq NaOH	281	#19	2 52	19 86	3 2	343	3 4	298	1 62	2 40	6 02	5 02
	282	#19	2 49	17 64			3 5	295	1 48	2 37	6 03	5 02
Citric Acid / 2 eq NaOH	283	#19	2 52	17 09	4 4	269	4 9	265	2 04	2 41	6 03	5 02
	284	#19	2 52	20 13			4 9	267	2 16	2 38	6 02	5 03
Sodium Citrate	285	#19	Missing Sample									
	286	#19	2 52	19 84	8 0	234	8 7	125	2 00	2 43	6 05	5 07
EGTA / 2 eq NaOH	287	#19	2 51	20 24	6 0	318	5 0	27	1 64	2 49	6 39	5 38
	288	#19	2 54	20 24			4 9	25	1 57	2 50	6 30	5 29
EGTA / 3 eq NaOH	289	#19	2 51	20 19	8 9	167	8 7	-43	1 56	2 48	6 36	5 34
	290	#19	2 49	20 24			8 7	-42	1 59	2 43	6 36	5 37
EGTA / 3 5 eq NaOH	291	#19	2 48	20 51	9 2	71	9 2	54	1 61	2 43	6 35	5 35
	292	#19	2 48	20 50			9 2	55	1 57	2 40	5 03	4 03
EGTA / 4 eq NaOH	293	#19	2 49	20 52	9 9	44	9 7	77	1 73	2 42	6 44	5 43
	294	#19	2 53	20 48			9 8	78	1 63	2 45	6 45	5 44

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Citric Acid	279	447 ± 26	27 ± 2	2 253 ± 404	351 ± 43	37	36	669	62	63	5 143
	280	441 ± 26	26 ± 2	1 915 ± 360	330 ± 40	36		660	64		4 694
Citric Acid / 1 eq NaOH	281	347 ± 20	40 ± 3	2 836 ± 491	289 ± 32	54	52	694	51	57 *	5 342
	282	351 ± 20	38 ± 3	1 782 ± 480	311 ± 38	50		647	63		4 205
Citric Acid / 2 eq NaOH	283	267 ± 15	57 ± 4	2 374 ± 423	324 ± 37	69	71	676	56	58	4 699
	284	219 ± 13	48 ± 3	2 225 ± 457	308 ± 40	73		628	61		4 850
Sodium Citrate	285						42 0			35 0	
	286	430 ± 25	31 ± 2	1 908 ± 462	103 ± 22	42		692	35		2 778
EGTA / 2 eq NaOH	287	257 ± 15	50 ± 3	2,314 ± 361	211 ± 32	69	69	697	48	42	4 173
	288	250 ± 14	52 ± 3	2 731 ± 424	146 ± 22	70		705	35		4 008
EGTA / 3 eq NaOH	289	275 ± 16	50 ± 3	4 104 ± 564	146 ± 22	67	67	716	26	31	5 393
	290	283 ± 16	50 ± 4	3 176 ± 421	173 ± 24	66		727	35		4 714
EGTA / 3 5 eq NaOH	291	316 ± 18	56 ± 4	3,549 ± 488	122 ± 19	67	64	822	26	27	4 652
	292	319 ± 18	45 ± 3	3 902 ± 415	140 ± 32	62		747	28		5 235
EGTA / 4 eq NaOH	293	294 ± 17	44 ± 3	2 564 ± 415	95 ± 16	62	64	687	27	28	3 413
	294	292 ± 17	50 ± 3	2 726 ± 484	108 ± 22	65		733	28		3 679

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RIP (70 C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
Na2EDTA	295	#20	2 53	19 39	4 9	268	5 6	-42	1 54	2 37	5 91	4 90
	296	#20	2 52	19 39			5 6	-44	1 71	2 37	6 25	5 25
Na2EDTA / 1 eq NaOH	297	#20	2 48	19 75	8 1	171	8 3	87	1 82	2 40	6 17	5 19
	298	#20	2 51	19 65			8 3	88	1 79	2 36	6 16	5 19
Na2EDTA / 2 eq NaOH	299	#20	2 51	19 77	11 0	57	10 2	62	1 63	2 40	6 16	5 16
	300	#20	2 54	19 76			10 3	64	1 77	2 42	6 20	5 20
DTPA / 2 eq NaOH	301	#20	2 49	19 87	3 3	340	3 8	3	1 74	2 38	6 27	5 25
	302	#20	2 48	19 82			3 8	5	1 86	2 39	6 19	5 18
DTPA / 3 eq NaOH	303	#20	2 54	19 90	4 9	286	6 7	32	1 77	2 43	5 93	4 92
	304	#20	2 47	19 85			6 6	31	1 78	2 38	6 14	5 14
DTPA / 4 eq NaOH	305	#20	2 46	19 93	8 9	123	8 9	16	1 68	2 40	6 18	5 16
	306	#20	Missing Sample									
DTPA / 5 eq NaOH	307	#20	2 52	18 91	10 4	34	10 3	57	1 62	2 47	6 20	5 20
	308	#20	2 50	19 92			10 3	57	1 58	2 74	6 13	5 12
DI Water	309	#20	2 47	19 38	5 7	70	7 9	179	1 85	2 44	5 91	4 94
	310	#20	2 49	19 33			7 9	174	2 10	2 47	5 98	4 98

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Na2EDTA	295	197 ± 11	62 ± 4	2 121 ± 629	278 ± 32	79	75	724	57	57	4 483
	296	202 ± 12	45 ± 3	2,249 ± 536	283 ± 43	71		578	56		4 611
Na2EDTA / 1 eq NaOH	297	254 ± 15	58 ± 4	2 298 ± 423	179 ± 27	73	72	753	44	42	3 838
	298	249 ± 14	55 ± 4	2 830 ± 512	184 ± 24	72		715	39		4 387
Na2EDTA / 2 eq NaOH	299	282 ± 16	45 ± 3	3 301 ± 387	159 ± 24	63	64	670	32	33	4 673
	300	283 ± 16	50 ± 3	3 159 ± 378	173 ± 24	66		706	35		4 623
DTPA / 2 eq NaOH	301	195 ± 11	57 ± 4	3 903 ± 625	432 ± 54	79	79	691	54	62	7 659
	302	207 ± 12	60 ± 4	2 029 ± 556	405 ± 45	79		726	70		5 531
DTPA / 3 eq NaOH	303	241 ± 14	60 ± 4	2 540 ± 427	211 ± 30	75	75	758	46	48	4 359
	304	227 ± 13	55 ± 4	2 041 ± 419	197 ± 32	75		709	50		3 766
DTPA / 4 eq NaOH	305	295 ± 17	51 ± 3	2 402 ± 620	378 ± 22	66	66 0	748	64	64 0	5 756
	306										
DTPA / 5 eq NaOH	307	283 ± 16	53 ± 4	3 212 ± 594	195 ± 27	66	66	717	36	38	4 808
	308	280 ± 16	48 ± 3	2 662 ± 442	178 ± 24	65		702	40		4 227
DI Water	309	684 ± 40	0 8 ± 0 3	6 340 ± 587	8 ± _	1	1	691	1	1	6 408
	310	700 ± 41	1 1 ± 0 3	6 128 ± 510	8 ± _	1		709	1		6 194

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RIT (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
2 M Sodium Carbonate	311	#21	2 51	23 12	11 4	33	10 8	177	1 45	2 70	7 24	6 14
	312	#21	2 52	23 04			10 8	170	1 44	2 70	7 18	6 10
Sodium Carbonate	313	#21	2 51	19 15	11 2	176	10 0	24	1 69	2 38	6 15	5 22
	314	#21	2 50	19 69			10 1	25	1 81	2 39	6 15	5 28
Sodium Bicarbonate	315	#21	2 49	19 72	8 8	396	8 5	84	1 65	2 44	6 27	5 24
	316	#21	2 52	19 69			8 6	84	1 71	2 45	6 27	5 24
0 05 M NTA / 1 eq NaOH	317	#21	2 51	19 65	2 6	386	5 7	126	1 87	2 44	6 22	5 28
	318	#21	2 47	19 61			5 7	140	1 86	2 37	6 18	5 24
0 08 M NTA / 1 5 eq NaOH	319	#21	2 50	19 67	2 8	386	5 4	90	1 66	2 37	6 46	5 51
	320	#21	2 51	19 64			5 4	40	1 85	2 39	6 52	5 57
NTA / 2 eq NaOH	321	#21	2 52	19 74	3 6	344	6 3	38	2 44	2 44	6 43	5 50
	322	#21	2 53	19 71			6 3	21	2 48	2 45	6 44	5 49
NTA / 2 5 eq NaOH	323	#21	2 53	19 78	8 6	178	7 3	95	2 74	2 46	6 45	5 48
	324	#21	2 50	19 77			7 4	93	2 65	2 45	6 39	5 43
NTA / 3 eq NaOH	325	#21	2 53	19 74	10 3	61	9 7	24	1 68	2 42	6 43	5 42
	326	#21	2 51	19 71			9 7	28	1 80	2 41	6 24	5 37

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
2 M Sodium Carbonate	311	496 ± 29	12 ± 1 1	3 749 ± 592	78 ± 14	21	20	618	19	18	4,542
	312	527 ± 31	11 ± 1 0	3 421 ± 526	70 ± 14	19		638	18		4 128
Sodium Carbonate	313	718 ± 42	2 8 ± 0 2	4 265 ± 557	16 ± 11	3	3	741	3	2	4 396
	314	737 ± 43	2 7 ± 0 2	4 697 ± 566	8 ± 8	3		759	2		4 764
Sodium Bicarbonate	315	698 ± 41	0 7 ± 0 3	3 705 ± 440	3 ± 1	1	1	704	1	1	3 733
	316	696 ± 41	*0 3* ±	4 372 ± 436	*8* ±	0		699	2		4 440
0 05 M NTA / 1 eq NaOH	317	281 ± 16	53 ± 3	2 919 ± 512	257 ± 24	68	69	723	47	51	5 062
	318	266 ± 15	56 ± 4	2 351 ± 465	270 ± 24	71		741	55		4 642
0 08 M NTA / 1 5 eq NaOH	319	231 ± 13	55 ± 4	2 159 ± 382	332 ± 27	73	75	696	62	56	4 964
	320	235 ± 14	63 ± 4	2 273 ± 396	227 ± 28	76		759	50		4 160
NTA / 2 eq NaOH	321	246 ± 14	60 ± 4	4 041 ± 506	275 ± 35	76	75	728	40	44	6 250
	322	236 ± 14	56 ± 4	3 229 ± 460	297 ± 40	75		683	48		5 599
NTA / 2 5 eq NaOH	323	282 ± 16	56 ± 3	2 277 ± 380	297 ± 30	71	71	726	59	58	4 629
	324	273 ± 16	56 ± 3	2 071 ± 335	259 ± 34	72		724	58		4 157
NTA / 3 eq NaOH	325	340 ± 20	50 ± 3	3 757 ± 657	92 ± 19	61	62	763	19	22	4 536
	326	302 ± 17	51 ± 3	4 628 ± 613	161 ± 22	64		726	25		5 966

Appendix D Data for Batch Experiments at 80 C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RH (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
Ascorbic Acid	327	#22	2 52	20 06	2 6	136	3 9	86	1 70	2 48	6 38	5 36
	328	#22	2 51	20 02			3 9	88	1 53	2 44	6 39	5 37
Dithionite	329	#22	2 52	19 35	3 1	286	4 0	92	1 75	2 48	6 34	5 32
	330	#22	2 50	19 65			4 0	111	1 45	2 46	6 28	5 19
Hydrogen Peroxide	331	#22	2 49	19 70	4 3	382	4 7	157	1 56	2 29	6 08	5 07
	332	#22	2 51	19 57			4 7	153	1 46	2 26	6 45	5 44
NaOCl	333	#22	2 52	21 59	10 9	625	6 9	114	1 17	2 47	6 59	5 50
	334	#22	2 52	21 59			7 1	114	1 40	2 48	6 53	5 43
Persulfate	335	#22	2 51	20 29	5 0	444	2 0	363	1 38	2 39	6 16	5 14
	336	#22	2 50	20 25			1 8	361	1 76	2 36	6 12	5 10
Na3Citrate / Ascorbic Acid	337	#22	2 49	20 07	5 2	25	5 4	177	1 88	2 42	6 69	5 66
	338	#22	2 52	20 04			5 4	182	1 92	2 46	6 61	5 58
Na3Citrate / Dithionite	339	#22	2 52	20 57	6 0	540	6 4	248	1 53	2 43	6 61	5 57
	340	#22	2 50	20 60			6 4	246	1 66	2 43	6 62	5 58
Na3Citrate / Hydrogen Peroxide	341	#22	2 50	20 19	7 5	240	8 3	28	2 22	2 35	6 53	5 50
	342	#22	2 52	20 34			8 2	26	2 11	2 38	6 59	5 56

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Ascorbic Acid	327	583 ± 34	7 6 ± 0 6	4 134 ± 493	40 ± 8	11	10	649	8	10	4 480
	328	623 ± 37	6 5 ± 0 7	4 616 ± 487	59 ± 18	9		680	11		5 134
Dithionite	329	715 ± 42	0 4 ± 0 2	5 098 ± 640	3 2 ± 1 4	1	1	718	1	0	5 125
	330	773 ± 45	0 5 ± 0 2	4 361 ± 621	1 9 ± 0 5	1		777	0		4 378
Hydrogen Peroxide	331	728 ± 43	*0 3* ±	5 313 ± 476	3 0 ± 0 6	0	0	731	1	1	5 339
	332	724 ± 43	*0 3* ±	4 901 ± 516	*5* ±	0		727	1		4 944
NaOCl	333	667 ± 39	3 7 ± 0 4	4 303 ± 392	62 ± 24	5	6	703	13	15	4 904
	334	657 ± 39	3 8 ± 0 3	4 067 ± 429	78 ± 40	6		694	17		4 820
Persulfate	335	289 ± 17	51 ± 3	3 864 ± 496	159 ± 27	66	66	749	29	25	5 299
	336	294 ± 17	52 ± 3	3 928 ± 456	108 ± 13	67		755	21		4 886
Na3Citrate / Ascorbic Acid	337	218 ± 13	51 ± 4	2 191 ± 405	178 ± 21	74	73	657	45	51	3 725
	338	242 ± 14	53 ± 4	1 738 ± 338	208 ± 27	72		694	56		3 510
Na3Citrate / Dithionite	339	260 ± 15	47 ± 3	2 045 ± 314	255 ± 22	67	67	682	57	57	4 334
	340	277 ± 16	48 ± 3	2 457 ± 363	290 ± 33	66		709	56		5 064
Na3Citrate / Hydrogen Peroxide	341	445 ± 26	31 ± 2	2 346 ± 477	175 ± 27	42	42	709	44	40	3 838
	342	458 ± 27	32 ± 2	3 477 ± 505	202 ± 32	42		732	37		5 209

Appendix D Data for Batch Experiments at 80 C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20 C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
Ascorbic Acid / 1 eq NaOH	343	#23	2 52	20 10	5 6	34	6 6	195	1 40	2 48	6 21	5 20
	344	#23	2 52	20 09			6 6	196	1 42	2 47	6 39	5 38
Na3Cit / 1 eq NaOH / Ascorbic Acid	345	#23	2 52	20 42	7 0	54	7 2	224	1 84	2 44	6 25	5 23
	346	#23	2 50	20 45			7 2	227	1 89	2 44	6 29	5 24
Na3Citrate / Persulfate	347	#23	2 52	20 70	7 7	419	5 0	9	1 55	2 44	6 39	5 36
	348	#23	2 49	20 72			5 1	5	1 53	2 39	6 35	5 33
0 3 M Na3Cit / 0 01 M NaHCO3 / 0 5 M Dithionite	349	#23	2 51	21 41	6 0	560	5 8	199	1 43	2 47	6 56	5 48
	350	#23	2 51	21 43			5 8	199	1 33	2 49	6 49	5 41
Na2EDTA / 2 eq NaOH / Ascorbic Acid	351	#23	2 50	20 56	7 6	99	7 4	228	1 52	2 38	6 45	5 42
	352	#23	2 50	20 57			7 4	229	1 53	2 38	6 36	5 34
Na2EDTA / 1 eq NaOH / Dithionite	353	#23	2 52	20 54	6 9	609	6 5	110	1 47	2 38	6 43	5 39
	354	#23	2 50	20 60			6 5	111	1 49	2 36	6 40	5 37
Na2EDTA / 1 eq NaOH / Hydrogen Peroxide	355	#23	2 49	20 58	8 5	169	7 5	49	2 19	2 37	6 23	5 21
	356	#23	2 53	20 57			7 5	46	1 88	2 41	6 28	5 25
Na2EDTA / 1 eq NaOH / Na2S2O8	357	#23	2 49	20 61	4 7	403	6 3	76	2 10	2 40	6 23	5 20
	358	#23	2 49	20 63			6 3	77	2 17	2 43	6 16	5 13

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
Ascorbic Acid / 1 eq NaOH	343	398 ± 23	31 ± 2	2 678 ± 466	76 ± 19	44	45	673	22	30	3 351
	344	413 ± 24	34 ± 2	4,241 ± 697	260 ± 36	45		713	38		6,531
Na3Cit / 1 eq NaOH / Ascorbic Acid	345	310 ± 18	46 ± 3	2 612 ± 483	172 ± 24	62	61	716	40	40	4 129
	346	318 ± 18	42 ± 3	3,278 ± 516	205 ± 21	60		692	39		5 105
Na3Citrate / Persulfate	347	211 ± 12	58 ± 4	2 607 ± 469	332 ± 33	77	77	737	58	57	5 616
	348	214 ± 12	56 ± 4	2 417 ± 486	277 ± 31	76		730	55		4 968
0 3 M Na3Cit / 0 01 M NaHCO3 / 0 5 M Dithionite	349	249 ± 14	54 ± 4	1,527 ± 489	457 ± 46	72	70	765	79	68 †	5 895
	350	287 ± 17	50 ± 3	2 249 ± 450	267 ± 32	67		767	57		4 814
Na2EDTA / 2 eq NaOH / Ascorbic Acid	351	207 ± 12	56 ± 4	2 987 ± 545	314 ± 32	77	77	715	53	59 *	5 835
	352	186 ± 11	53 ± 4	2 369 ± 430	405 ± 49	78		668	66		6 049
Na2EDTA / 1 eq NaOH / Dithionite	353	147 ± 8	58 ± 4	2,596 ± 483	316 ± 32	84	83	671	56	60	5 451
	354	167 ± 9	61 ± 4	2 318 ± 395	370 ± 49	83		722	64		5 687
Na2EDTA / 1 eq NaOH / Hydrogen Peroxide	355	308 ± 18	48 ± 3	4 097 ± 694	124 ± 16	65	62	732	24	28	5 194
	356	342 ± 20	46 ± 3	2 700 ± 536	127 ± 19	60		748	32		3 822
Na2EDTA / 1 eq NaOH / Na2S2O8	357	341 ± 20	43 ± 3	3 349 ± 582	227 ± 30	59	56	723	42	39	5 368
	358	441 ± 26	45 ± 3	4 028 ± 568	215 ± 22	53		841	36		5 941

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
DTPA / 3 37 eq NaOH	359	#24	2 51	20 43	8 0	180	7 4	103	1 34	2 40	6 86	5 84
	360	#24	2 51	20 48			7 4	100	1 52	2 38	6 17	5 15
DTPA / 4 50 eq NaOH / Ascorbic Acid	361	#24	2 52	20 47	8 0	209	6 9	202	1 46	2 35	6 12	5 09
	362	#24	2 52	20 59			6 9	200	1 39	2 30	7 09	6 07
DTPA / 4 32 eq NaOH / Na2S2O4	363	#24	2 50	20 88	8 1	677	6 5	163	1 44	2 33	7 06	6 03
	364	#24	2 50	20 78			6 5	161	1 44	2 31	7 46	6 43
DTPA / 3 5 eq NaOH / Hydrogen Peroxide	365	#24	2 52	20 40	8 0	221	7 4	18	2 09	2 31	6 19	5 17
	366	#24	2 51	20 52			7 4	10	2 19	2 38	7 23	6 20
DTPA /3 50 eq NaOH / Na2S2O8	367	#24	2 50	20 69	8 0	193	5 1	34	1 65	2 39	6 81	5 78
	368	#24	2 52	20 78			5 1	33	1 68	2 41	6 66	5 65
NTA / 3 33 eq NaOH / Ascorbic Acid	369	#24	2 50	20 50	8 0	173	7 0	190	1 51	2 37	6 29	5 27
	370	#24	2 50	20 46			7 0	190	1 50	2 36	6 33	5 31
NTA / 2 68 eq NaOH / Na2S2O4	371	#24	2 49	20 40	8 0	668	6 4	71	1 40	2 36	6 24	5 23
	372	#24	2 50	20 52			6 2	56	1 49	2 34	6 25	5 23
NTA / 2 22 eq NaOH / H2O2	373	#24	2 49	20 25	8 0	214	7 6	35	1 25	2 31	6 09	5 08
	374	#24	2 49	20 39			7 7	38	2 42	2 33	5 91	4 90

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
DTPA / 3 37 eq NaOH	359	228 ± 13	52 ± 4	2 520 ± 485	300 ± 32	72	74	692	55	52	5 197
	360	224 ± 13	58 ± 4	2 041 ± 398	193 ± 24	76		748	50		3 784
DTPA / 4 50 eq NaOH / Ascorbic Acid	361	223 ± 13	61 ± 4	2 134 ± 426	316 ± 32	77	76	776	62	53 †	5 001
	362	192 ± 11	52 ± 4	3 940 ± 551	317 ± 31	76		656	45		6 769
DTPA / 4 32 eq NaOH / Na2S2O4	363	102 ± 35	58 ± 4	1 870 ± 289	311 ± 32	90	86	631	65	68	4 707
	364	167 ± 10	57 ± 4	1 882 ± 340	400 ± 51	81		679	71		5 474
DTPA / 3 5 eq NaOH / Hydrogen Peroxide	365	269 ± 16	52 ± 4	2 139 ± 399	121 ± 18	70	70	723	37	38	3 194
	366	273 ± 16	53 ± 4	2 329 ± 383	148 ± 27	70		725	39		3 590
DTPA /3 50 eq NaOH / Na2S2O8	367	195 ± 11	53 ± 4	2 411 ± 426	227 ± 19	77	77	671	50	58	4 449
	368	207 ± 12	55 ± 4	1 677 ± 353	290 ± 32	77		700	66		4 276
NTA / 3 33 eq NaOH / Ascorbic Acid	369	228 ± 13	58 ± 4	1 680 ± 351	232 ± 21	75	75	755	60	61	3 788
	370	228 ± 13	55 ± 4	2 488 ± 415	362 ± 42	74		724	61		5 755
NTA / 2 68 eq NaOH / Na2S2O4	371	191 ± 11	65 ± 4	0 951 ± 601	286 ± 18	81	80	783	79	78	3,554
	372	198 ± 11	61 ± 4	1 221 ± 823	328 ± 37	79		752	77		4 202
NTA / 2 22 eq NaOH / H2O2	373	283 ± 16	47 ± 3	2 485 ± 366	162 ± 21	64	65	713	40	43	3 966
	374	294 ± 17	48 ± 3	2 298 ± 370	181 ± 24	67		711	46		3 872

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equibration with soil)	Eh of Lixiviant (after equibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
NTA / 2 18 eq NaOH / Na2S2O8	375	#25	2.51	20 47	8 2	423	2 9	283	1 53	2 45	6 46	5 44
	376	#25	2 53	20 37			2 9	280	1 52	2 43	6 15	5 11
NaHCO3/0 22 eq NaOH/Ascorbic Acid	377	#25	2 55	20 07	8 1	172	7 1	209	1 57	2 52	6 19	5 19
	378	#25	2 49	20 04			7 1	217	1 54	2 47	6 15	5 15
NaHCO3/0 12 eq NaOH/Na2S2O4	379	#25	2 49	20 22	8 0	-662	7 1	70	1 61	2 48	6 11	5 12
	380	#25	2 49	20 20			7 1	66	1 53	2 47	6 13	5 14
NaHCO3 / Na2S2O8	381	#25	2 50	20 25	8 0	288	2 2	327	1 45	2 40	5 79	4 79
	382	#25	2 49	20 25			2 2	325	1 45	2 39	6 15	5 13
EGTA / 3 27 eq NaOH / Ascorbic Acid	383	#25	2 50	20 51	8 0	202	6 2	167	1 34	2 44	6 39	5 51
	384	#25	2 50	20 47			6 2	168	1 34	2 44	5 94	4 91
EGTA / 2 89 eq NaOH / Na2S2O4	385	#25	2 51	20 65	8 0	666	6 4	32	1 32	2 44	6 91	5 90
	386	#25	2 49	20 70			6 4	39	1 33	2 44	6 67	5 65
EGTA / 2 30 eq NaOH/Na2S2O8	387	#25	2 49	20 66	8 0	261	2 9	207	1 55	2 46	6 02	5 00
	388	#25	2 50	20 70			2 9	205	1 65	2 48	5 97	4 95
Na3Cit / 0 53 eq NaOH / Na2S2O4	389	#25	2 50	20 49	7 8	660	6 7	105	1 31	2 42	5 99	4 97
	390	#25	2 48	20 51			6 7	141	1 40	2 40	5 95	4 93

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
NTA / 2 18 eq NaOH / Na2S2O8	375	481 ± 28	22 ± 2	2,545 ± 409	240 ± 24	31	32	678	49	59	4 694
	376	501 ± 29	24 ± 2	2 222 ± 429	435 ± 49	32		716	69		6 126
NaHCO3/0 22 eq NaOH/Ascorbic Acid	377	435 ± 25	32 ± 2	2 870 ± 485	137 ± 21	42	42	711	32	28	4 053
	378	428 ± 25	30 ± 2	3 841 ± 532	127 ± 18	42		694	25		4 967
NaHCO3/0 12 eq NaOH/Na2S2O4	379	677 ± 40	1 1 ± 0 2	4 596 ± 633	8 1 ± 3 0	2	1	687	2	2	4 668
	380	682 ± 40	0 9 ± 0 3	3 796 ± 535	6 9 ± 0 8	1		690	2		3 858
NaHCO3 / Na2S2O8	381	362 ± 21	40 ± 3	3 244 ± 551	62 ± 13	54	54	725	16	24	3 807
	382	367 ± 21	41 ± 3	2 413 ± 499	116 ± 13	54		738	33		3 462
EGTA / 3 27 eq NaOH / Ascorbic Acid	383	252 ± 14	65 ± 4	2 959 ± 447	303 ± 33	74	76	829	51	53	5 648
	384	227 ± 13	64 ± 4	2 666 ± 404	298 ± 31	77		819	54		5 423
EGTA / 2 89 eq NaOH / Na2S2O4	385	166 ± 10	57 ± 4	1 909 ± 254	305 ± 30	81	81	681	63	70 *	4 665
	386	178 ± 10	61 ± 4	1 709 ± 324	467 ± 58	81		738	76		5 999
EGTA / 2 30 eq NaOH/Na2S2O8	387	238 ± 14	55 ± 4	2 499 ± 501	240 ± 25	74	72	746	51	48	4 717
	388	248 ± 14	50 ± 3	2 882 ± 467	231 ± 21	71		709	46		5 010
Na3Cit / 0 53 eq NaOH / Na2S2O4	389	314 ± 18	40 ± 3	2 057 ± 422	214 ± 22	58	61	684	52	53	4 037
	390	289 ± 17	46 ± 3	2 485 ± 412	268 ± 33	64		717	54		4 978

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20°C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
NaHCO3 / 0 015 eq NaOH / 3% H2O2	391	#26	2 52	19 15	8 1	175	9 4	146	1 29	2 31	5 86	4 87
	392	#26	2 50	19 38			8 8	179	1 31	2 27	6 06	5 06
EGTA / 2 30 eq NaOH / 3% H2O2	393	#26	2 50	20 32	8 0	192	4 3	122	1 32	2 25	5 92	4 96
	394	#26	2 50	20 33			4 3	132	1 35	2 43	6 00	5 00
1% NaOCl / 0 40 eq H2SO4	395	#26	2 50	20 25	8 1	889	4 8	288	1 33	2 38	6 03	5 03
	396	#26	2 52	20 27			4 9	292	1 38	2 38	6 04	5 04
Na3Citrate / 0 76 eq H2SO4 / 1% NaOCl	397	#26	2 51	20 62	8 1	702	9 1	90	1 49	2 43	6 15	5 14
	398	#26	2 51	20 68			9 2	94	1 48	2 45	6 09	5 08
Na2EDTA / 0 83 eq NaOH / 1% NaOCl	399	#26	2 52	20 53	8 2	359	8 7	50	1 40	2 42	5 96	4 94
	400	#26	2 50	20 52			8 1	72	1 41	2 41	6 95	5 93
DTPA / 4 01 eq NaOH / 1% NaOCl	401	#26	2 52	20 65	8 2	47	8 3	23	1 47	2 44	6 02	5 00
	402	#26	2 51	20 64			7 9	35	1 46	2 45	6 01	4 99
NTA / 2 20 eq NaOH / 1% NaOCl	403	#26	2 50	20 52	8 1	189	8 0	70	1 47	2 41	6 02	5 00
	404	#26	2 50	20 44			8 2	67	1 50	2 41	5 95	4 93
NaHCO3 / 0 37 eq H2SO4 / 1% NaOCl	405	#26	2 49	20 40	8 1	898	8 8	153	1 33	2 38	5 89	4 89
	406	#26	2 51	20 41			8 1	176	1 38	2 39	5 91	4 91

Appendix D Data for Batch Experiments at 80°C

Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	↓ Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	↓ Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
NaHCO3 / 0 015 eq NaOH / 3% H2O2	391	765 ± 45	0 4 ± _	3 857 ± 645	5 4 ± 2 8	0	0	768	1	1	3 903
	392	746 ± 44	0 4 ± _	4 681 ± 595	4 2 ± 0 7	0		749	1		4 717
EGTA / 2 30 eq NaOH / 3% H2O2	393	256 ± 15	47 ± 3	2 281 ± 474	219 ± 23	67	68	683	50	46	4 273
	394	242 ± 14	49 ± 3	3 122 ± 554	216 ± 27	69		688	41		5 090
1% NaOCl / 0 40 eq H2SO4	395	739 ± 43	3 5 ± 0 6	5 998 ± 629	32 ± 8	4	5	771	5	5	6 288
	396	713 ± 42	3 6 ± 0 6	5 681 ± 610	37 ± 5	5		745	6		6 014
Na3Citrate / 0 76 eq H2SO4 / 1% NaOCl	397	464 ± 27	30 ± 2	3 412 ± 449	164 ± 32	40	41	738	33	37	4 910
	398	431 ± 25	30 ± 2	3 002 ± 303	197 ± 35	42		706	40		4 811
Na2EDTA / 0 83 eq NaOH / 1% NaOCl	399	245 ± 14	51 ± 3	3 514 ± 506	127 ± 11	70	68	712	27	29	4 676
	400	292 ± 17	52 ± 3	2 818 ± 445	135 ± 15	66		758	32		4 027
DTPA / 4 01 eq NaOH / 1% NaOCl	401	265 ± 15	52 ± 4	2 026 ± 303	219 ± 16	69	70	742	54	54	4 036
	402	230 ± 13	48 ± 3	2 290 ± 417	259 ± 37	71		672	55		4 674
NTA / 2 20 eq NaOH / 1% NaOCl	403	259 ± 15	40 ± 3	3 859 ± 624	205 ± 14	63	62	627	35	36	5 745
	404	280 ± 16	39 ± 3	3 284 ± 588	189 ± 21	60		637	37		5 012
NaHCO3 / 0 37 eq H2SO4 / 1% NaOCl	405	741 ± 43	2 4 ± 0 2	3 580 ± 657	40 ± 6	3	3	763	10	8	3 950
	406	739 ± 44	2 3 ± 0 4	4 670 ± 681	31 ± 4	3		760	6		4 953

Appendix D Data for Batch Experiments at 80°C

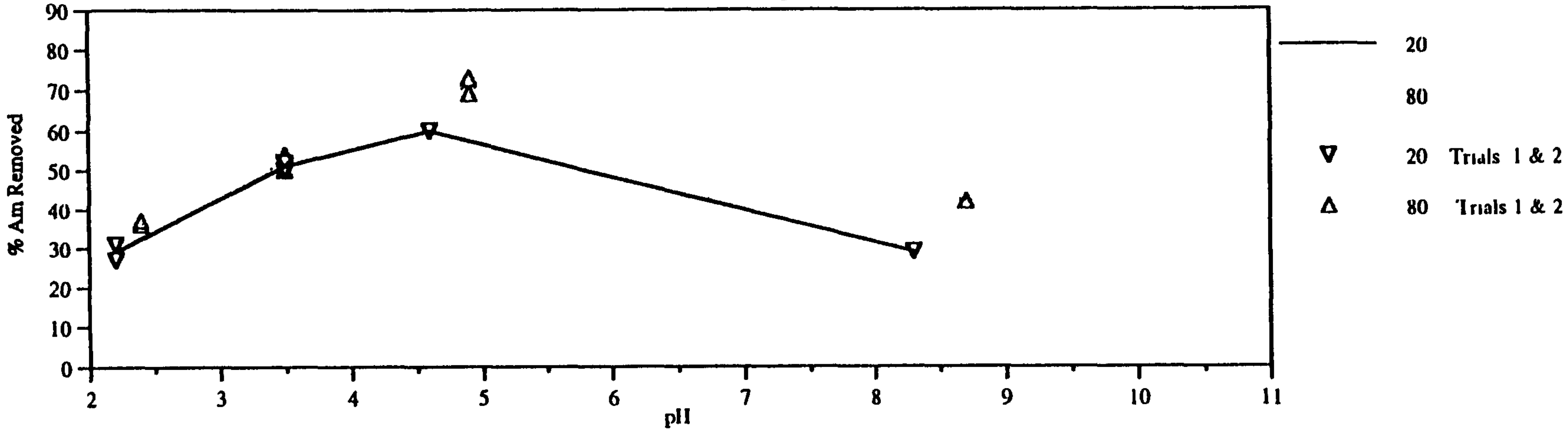
Leaching Solution (Lixiviant)	Sample No	Experiment No 94 RFP (20 C)	Grams of soil	Grams of Lixiviant	pH of Lixiviant	Eh of Lixiviant	pH of Lixiviant (after equilibration with soil)	Eh of Lixiviant (after equilibration with soil)	Grams of Liquid Associated with Wet Soil	Grams of dry soil	Grams of Extractant + Aq Soln (to prevent evap)	Grams of extractant in lq analyzed
EGTA / 2 38 eq H2SO4 / 1% NaOCl	407	#26	2 50	20 53	8 2	565	7 2	65	1 82	2 18	6 67	5 64
	408	#26	2 50	20 52			7 2	66	1 63	2 46	6 60	5 57

Appendix D Data for Batch Experiments at 80°C

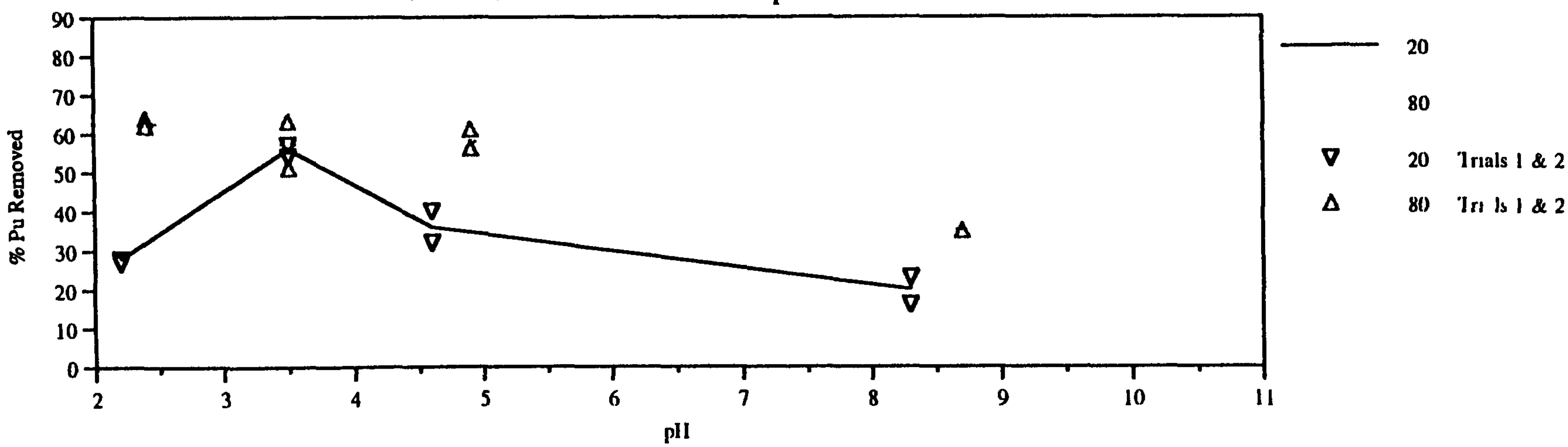
Leaching Solution (Lixiviant)	Sample No	Am 241 in Dry Soil (pCi/g)	Am 241 in Liquid (pCi/g)	Pu 239 in Dry Soil (pCi/g)	Pu 239 in Liquid (pCi/g)	% Am Removed	Average Am Removed (%)	Total Am 241 in Soil Used (pCi/g)	% Pu Removed	Average Pu Removed (%)	Total Pu 239 in Soil Used (pCi/g)
EGTA / 2 38 eq H2SO4 / 1% NaOCl	407	258 ± 15	46 ± 3	2,532 ± 460	170 ± 14	67	67	664	41	43	4 033
	408	274 ± 16	50 ± 3	2 882 ± 484	237 ± 27	67		721	46		4 999

Citric Acid System

% Am Removal as a Function of pH at 20° and 80°



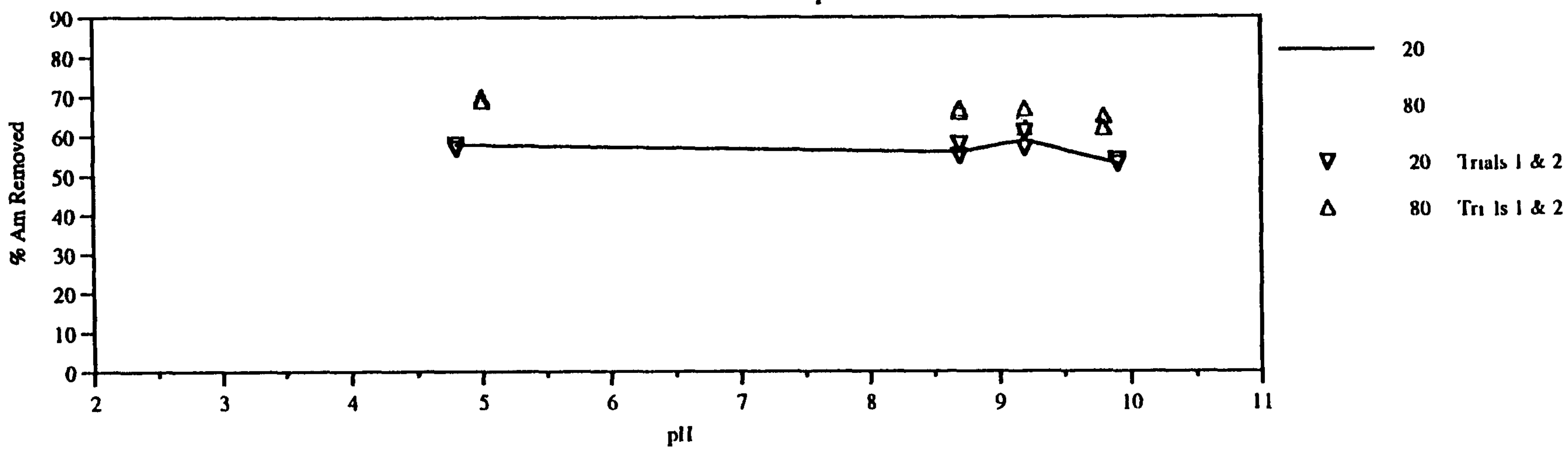
% Pu Removal as a Function of pH at 20 and 80°



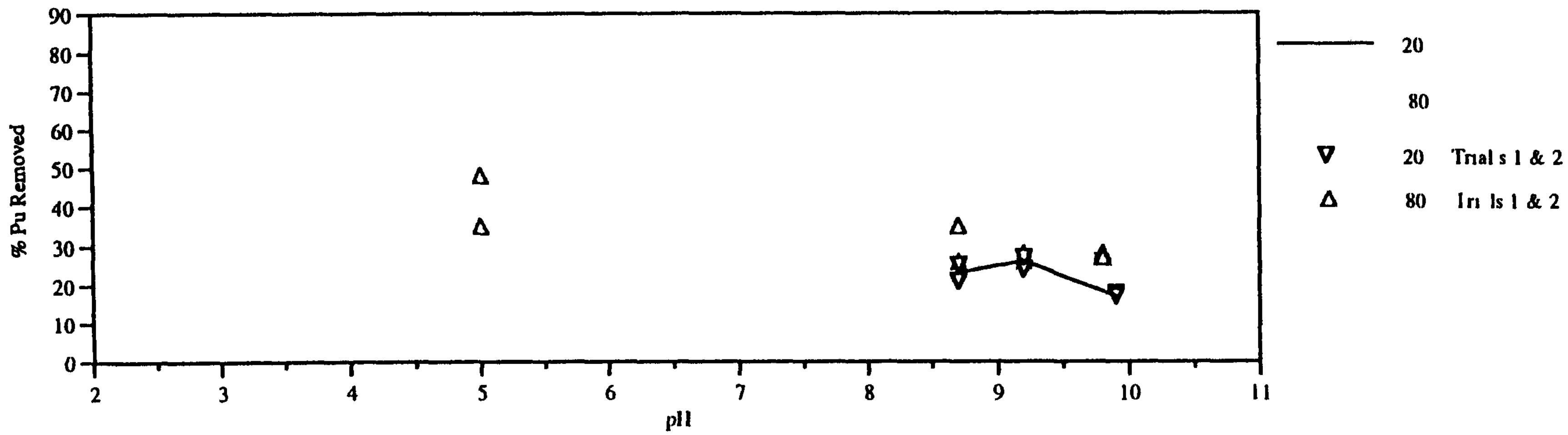
E 2

EGTA System

% Am Removal as a Function of pH at 20° and 80°



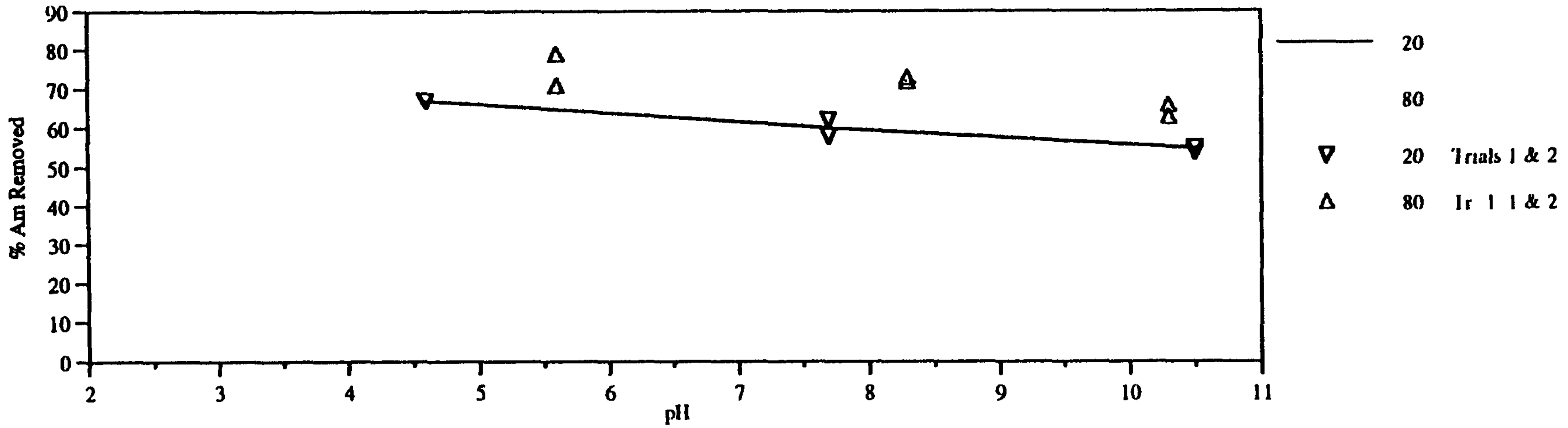
% Pu Removal as a Function of pH at 20° and 80



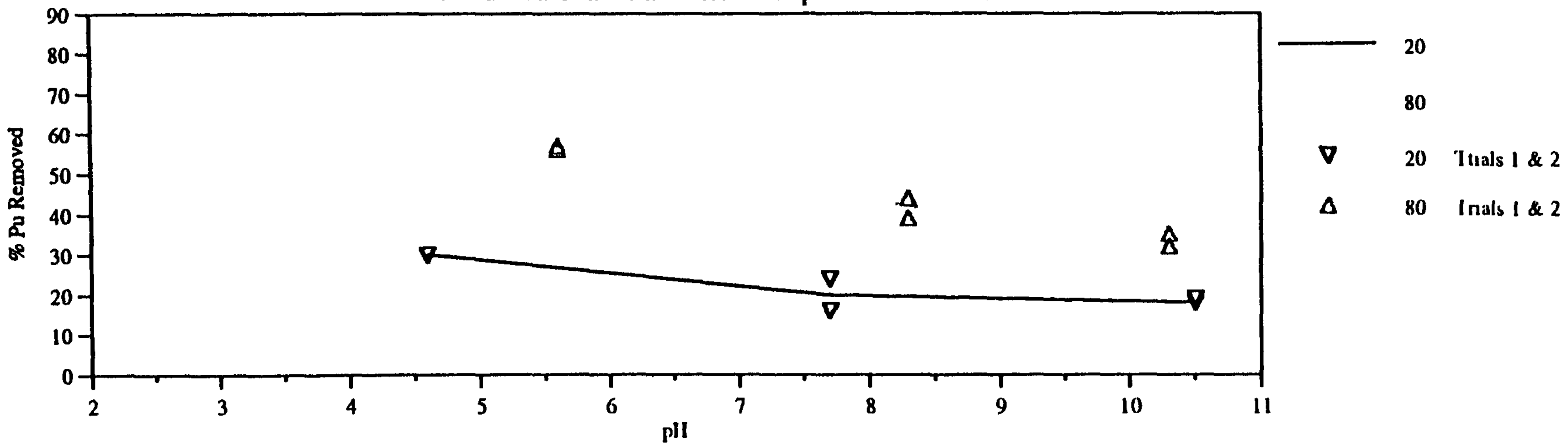
E 3

EDTA System

% Am Removal as a Function of pH at 20° and 80°

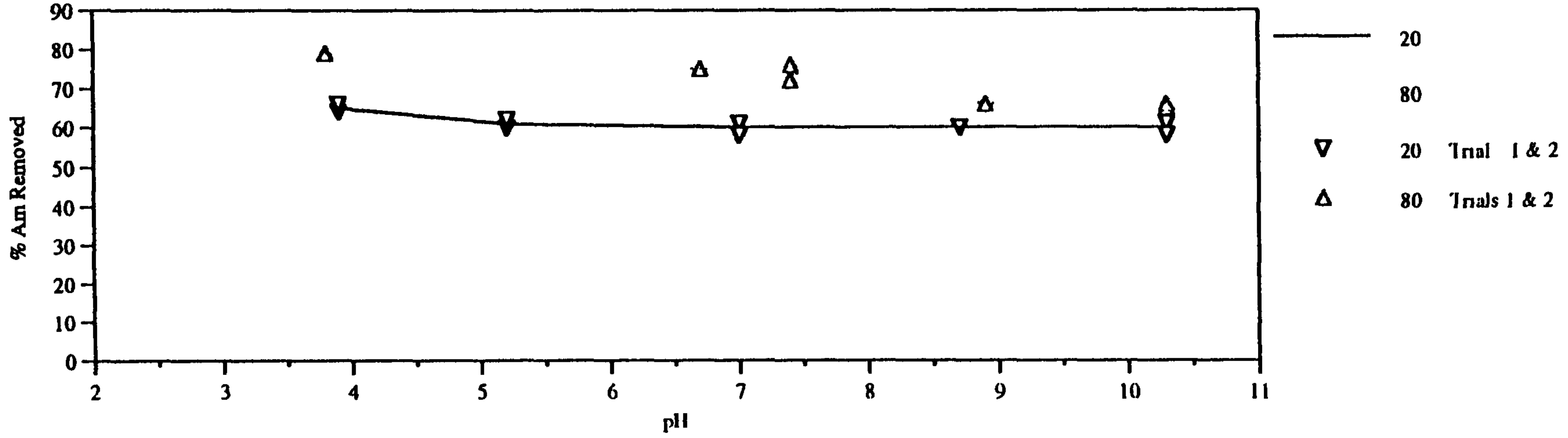


% Pu Removal as a Function of pH at 20 and 80

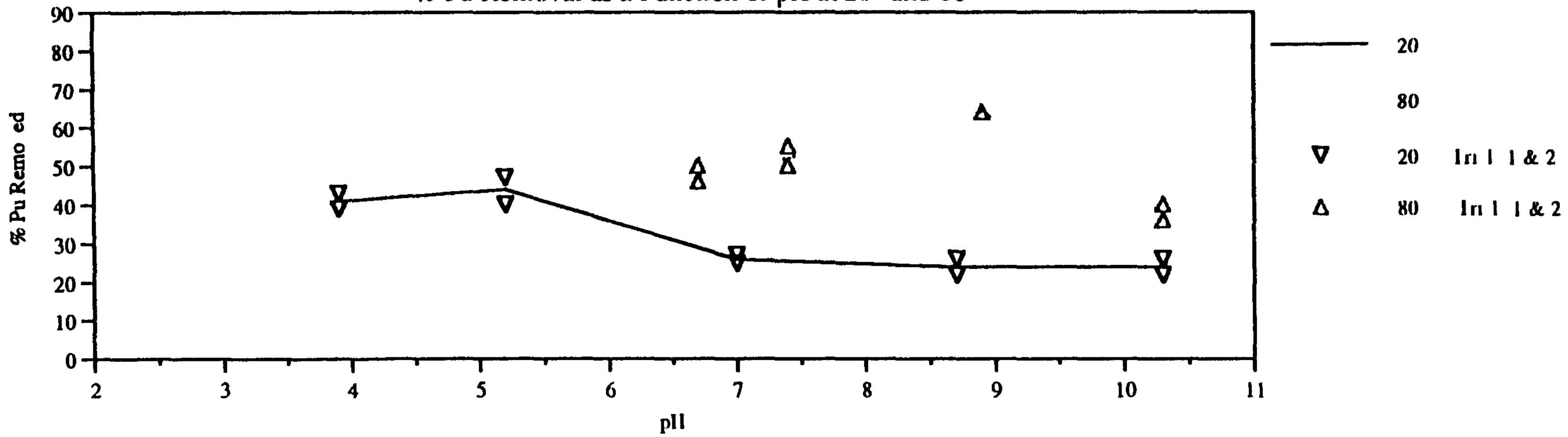


DTPA System

% Am Removal as a Function of pH at 20 and 80°

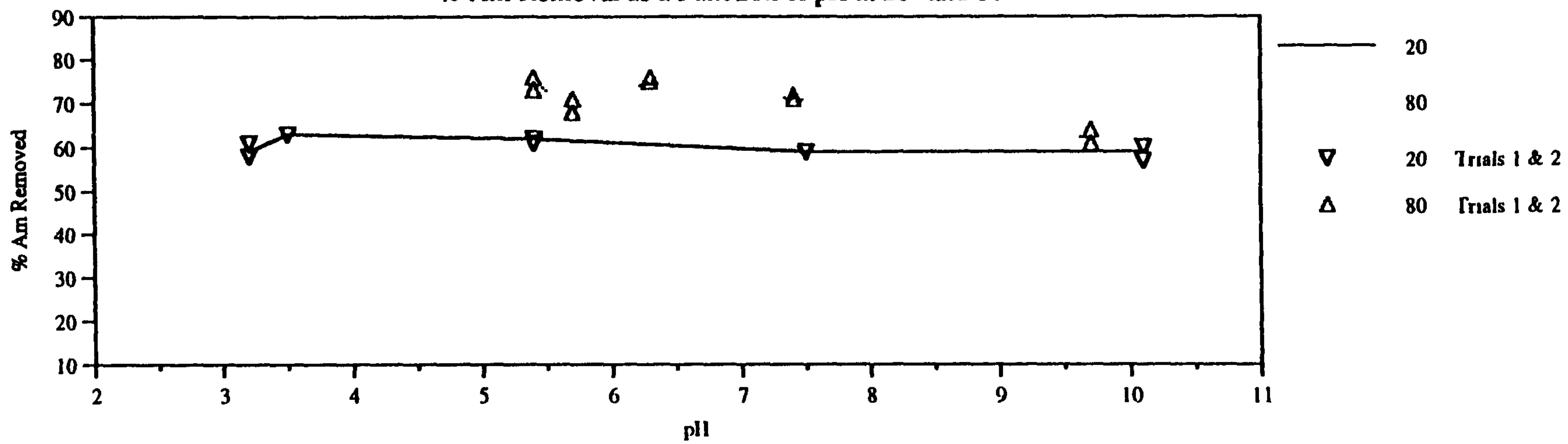


% Pu Removal as a Function of pH at 20° and 80°

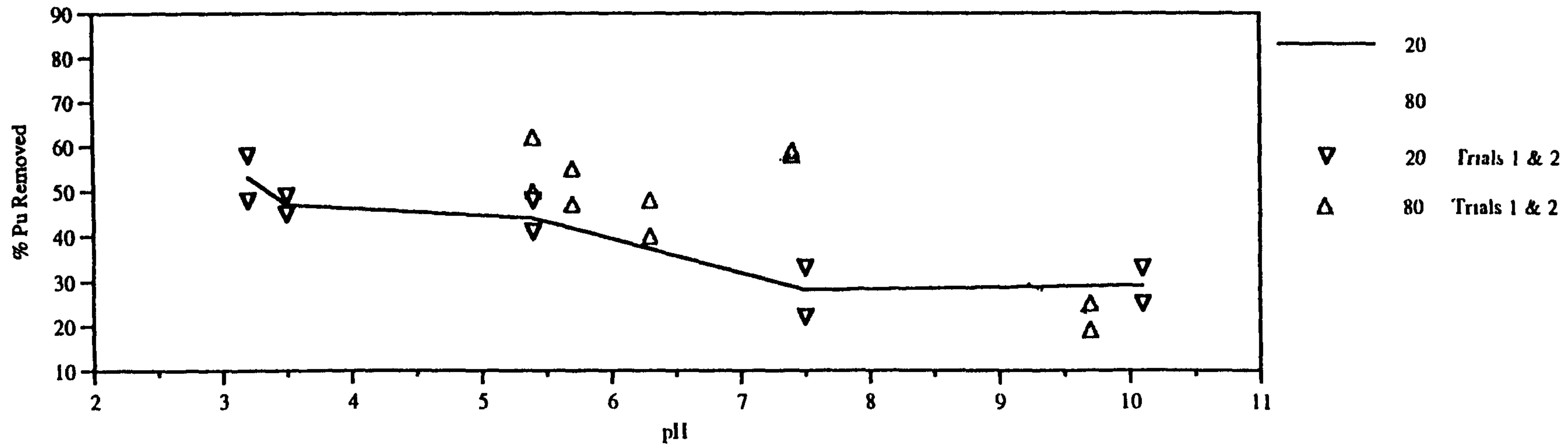


NTA System

% Am Removal as a Function of pH at 20° and 80°

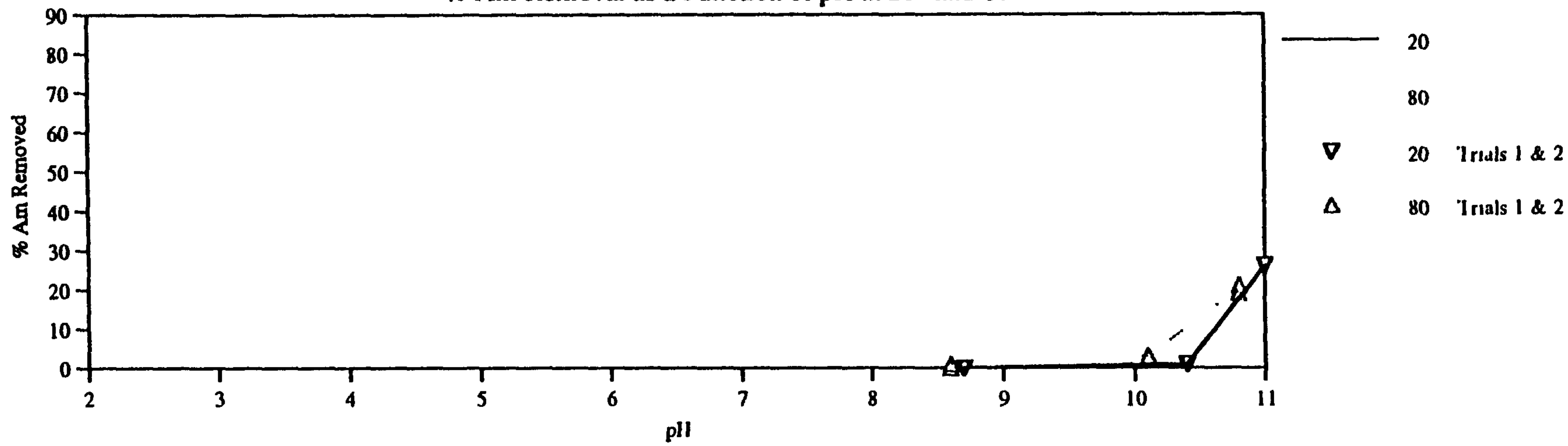


% Pu Removal as a Function of pH at 20 and 80°

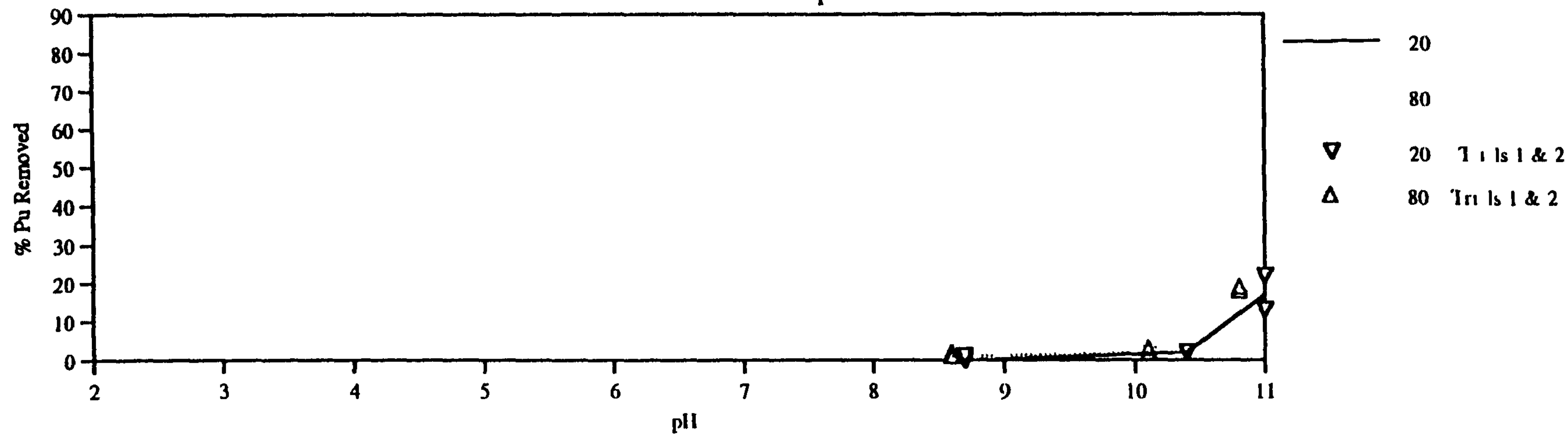


Carbonate System

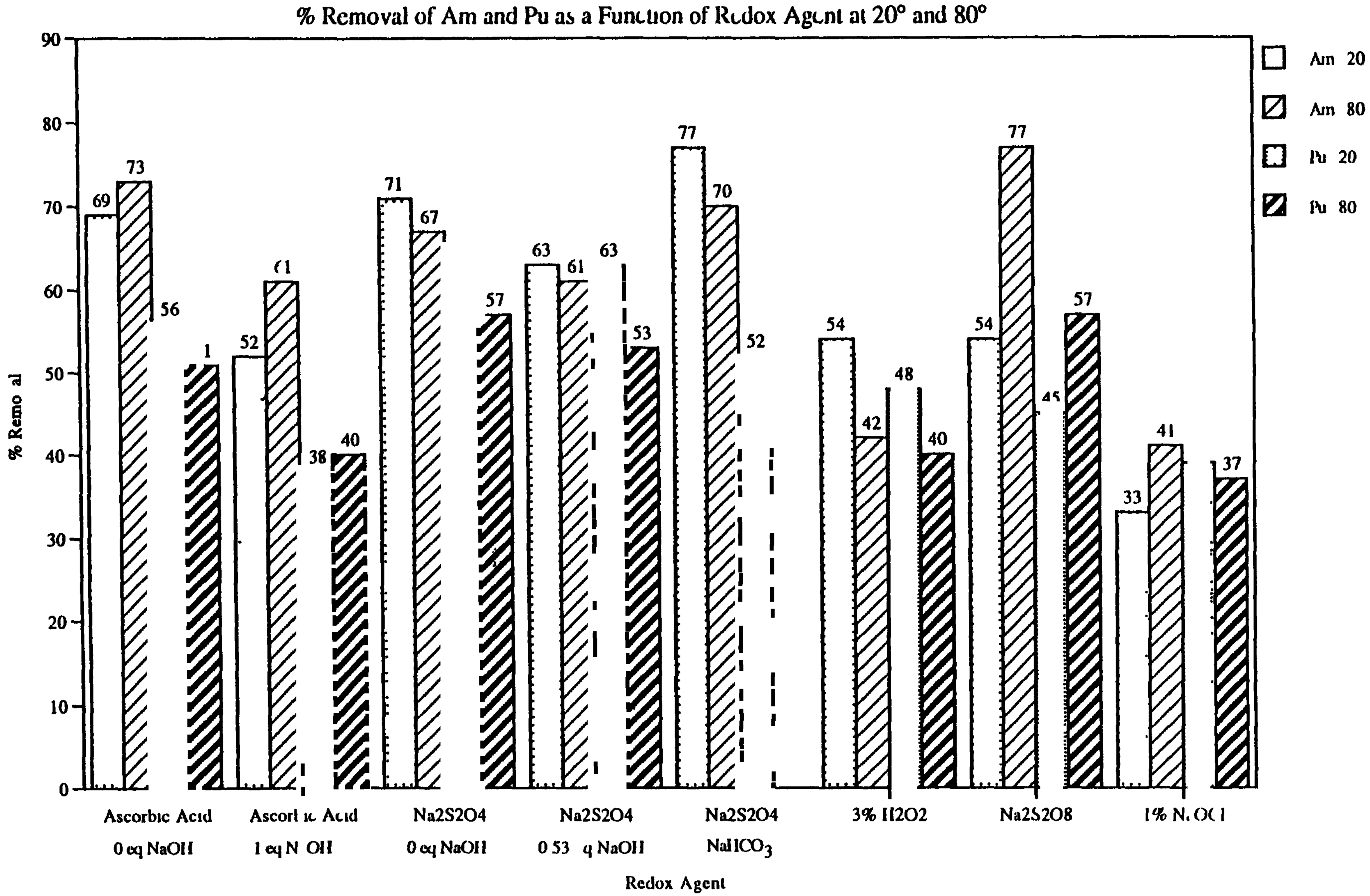
% Am Removal as a Function of pH at 20° and 80°



% Pu Removal as a Function of pH at 20 and 80

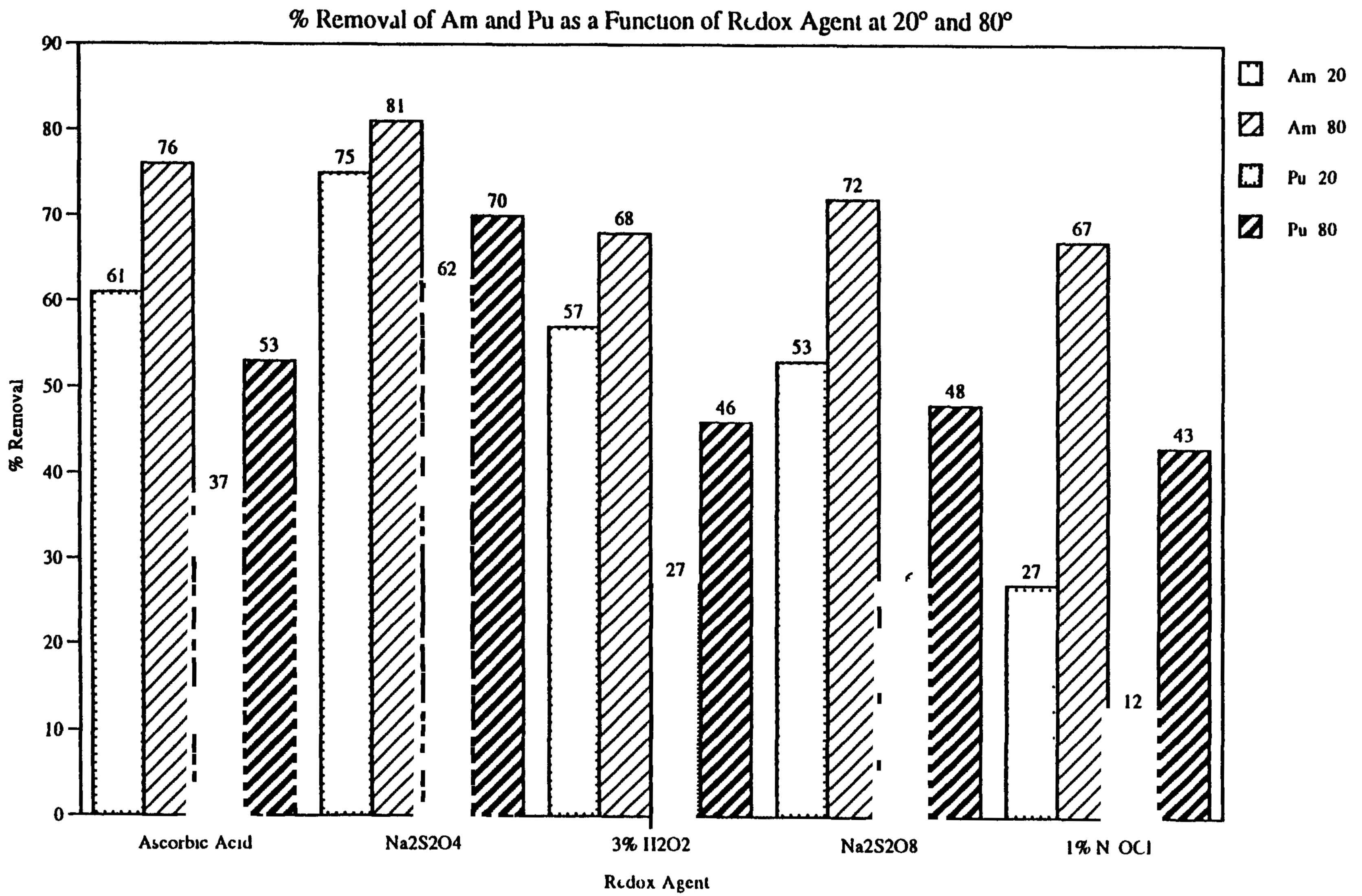


Na_3Citrate System



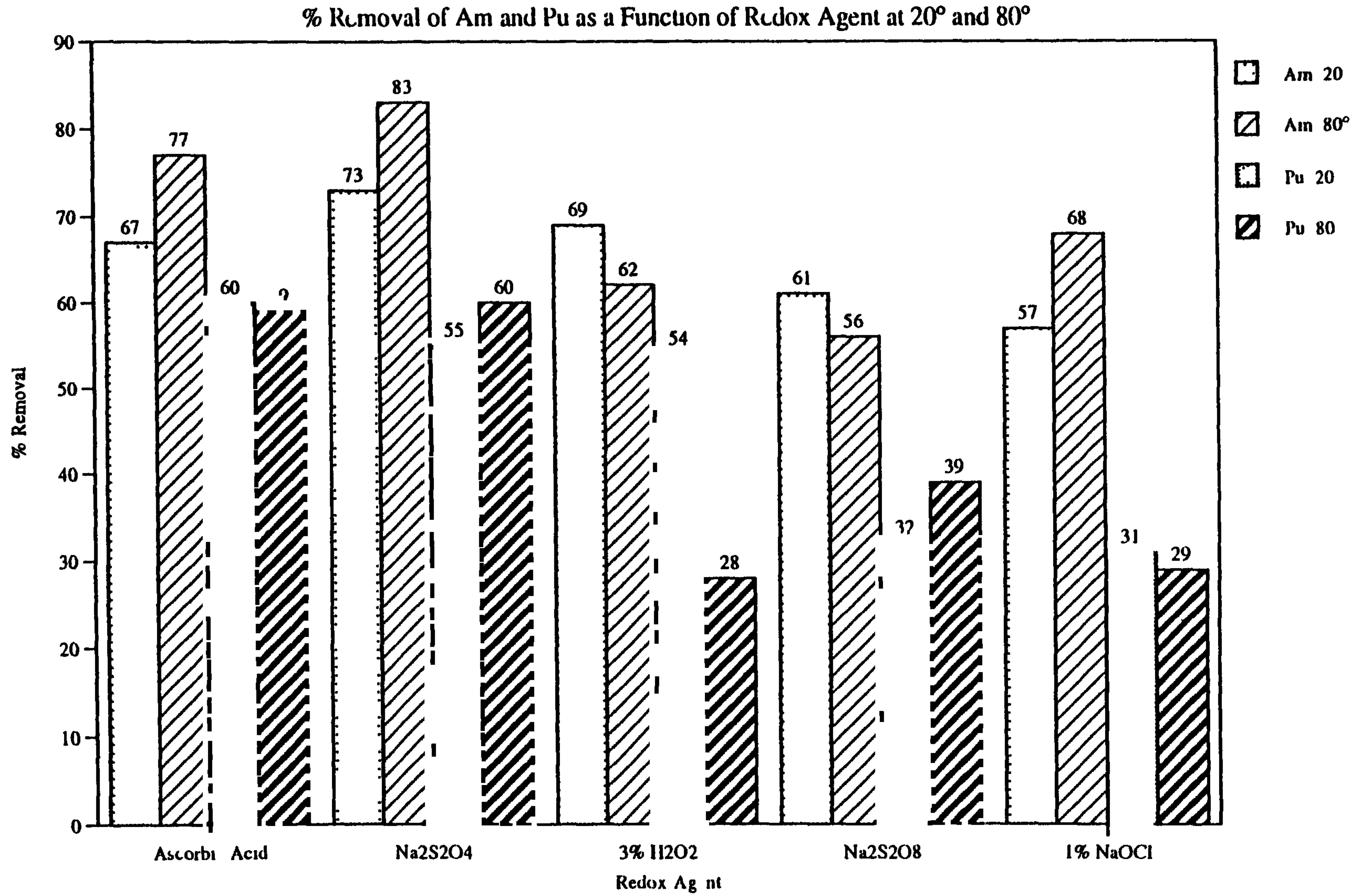
F 2

EGTA System



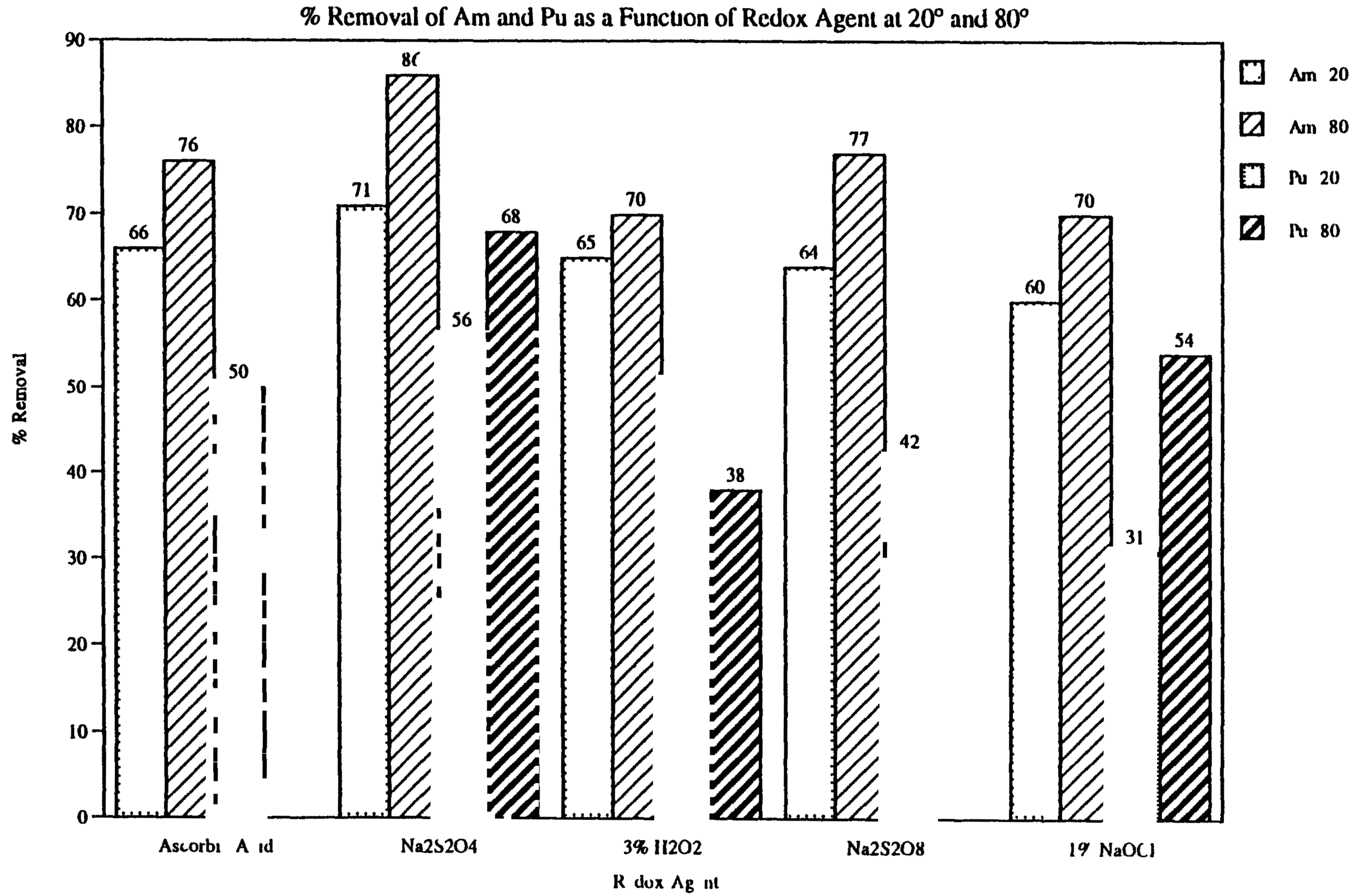
F 3

EDTA System



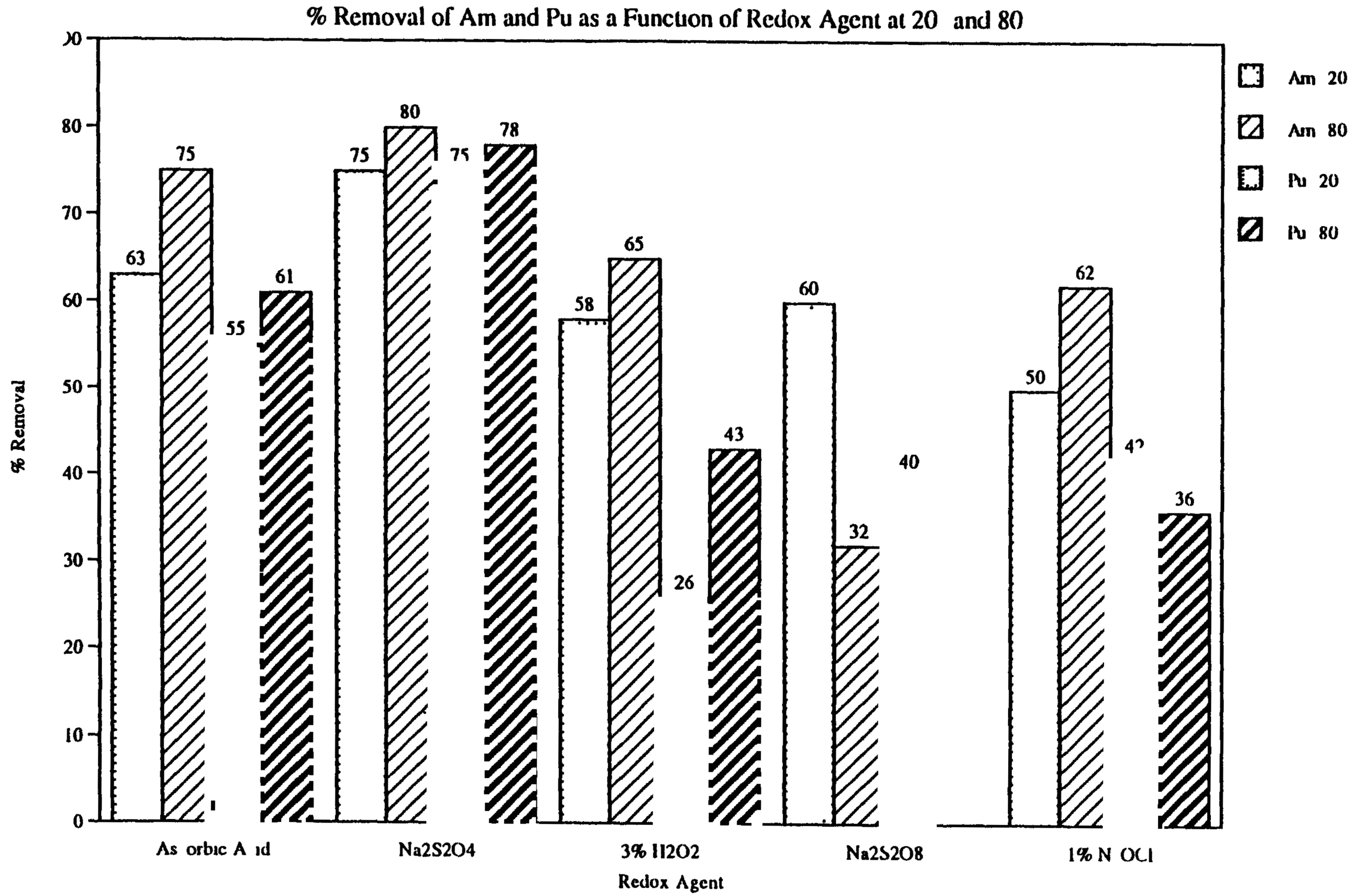
% Removal of Am and Pu as a Function of Redox Agent at 20° and 80°
% Removal
0
10
20
30
40
50
60
70
80
90
67
77
60
?
73
83
55
60
69
62
54
28
61
56
32
39
57
68
31
29
Am 20
Am 80°
Pu 20
Pu 80
Ascorbi Acid
Na2S2O4
3% H2O2
Na2S2O8
1% NaOCl
Redox Ag nt

DTPA System



F 5

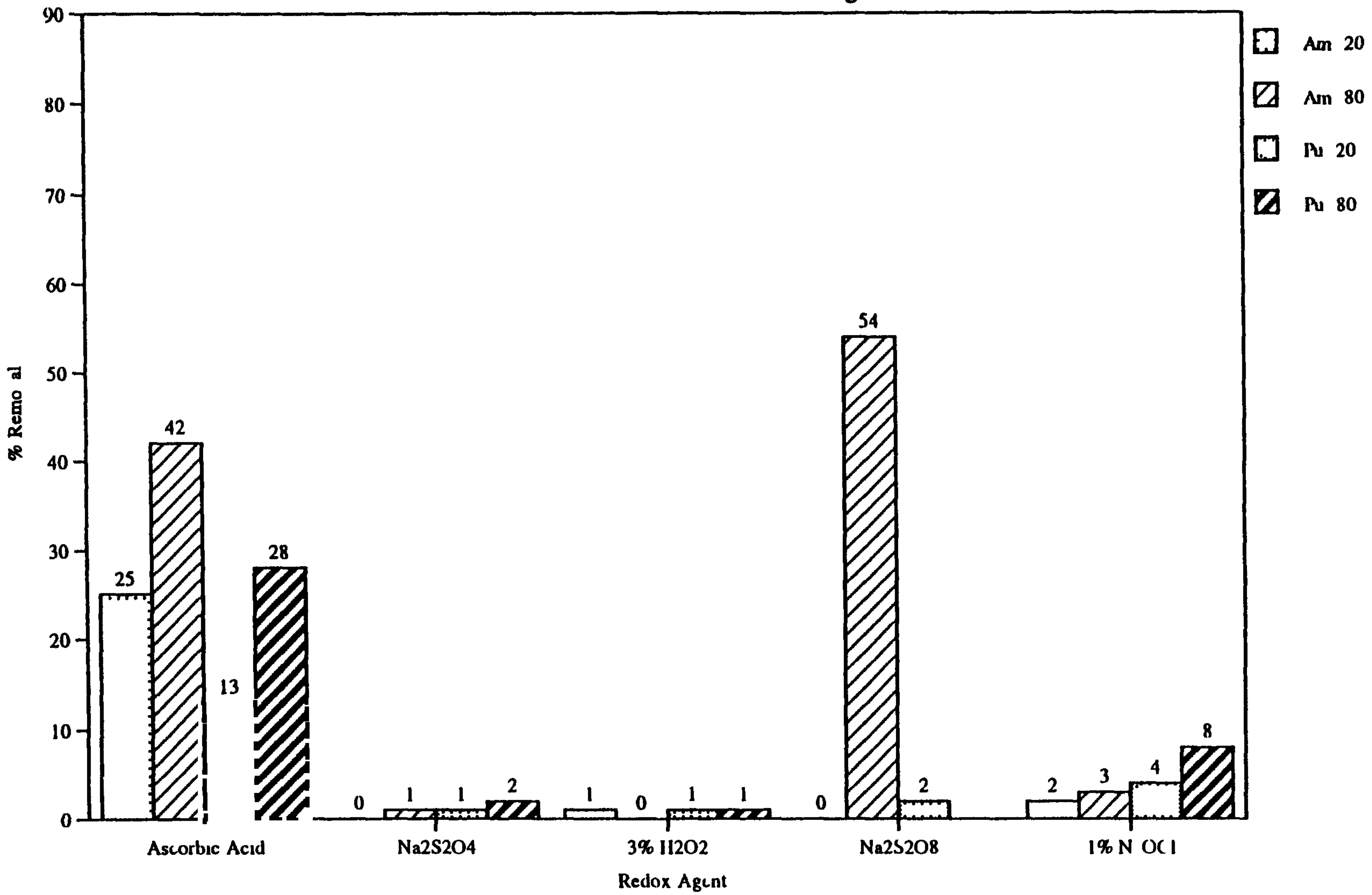
NTA System



F 6

Bicarbonate System

% Removal of Am and Pu as a Function of Redox Agent at 20° and 80°

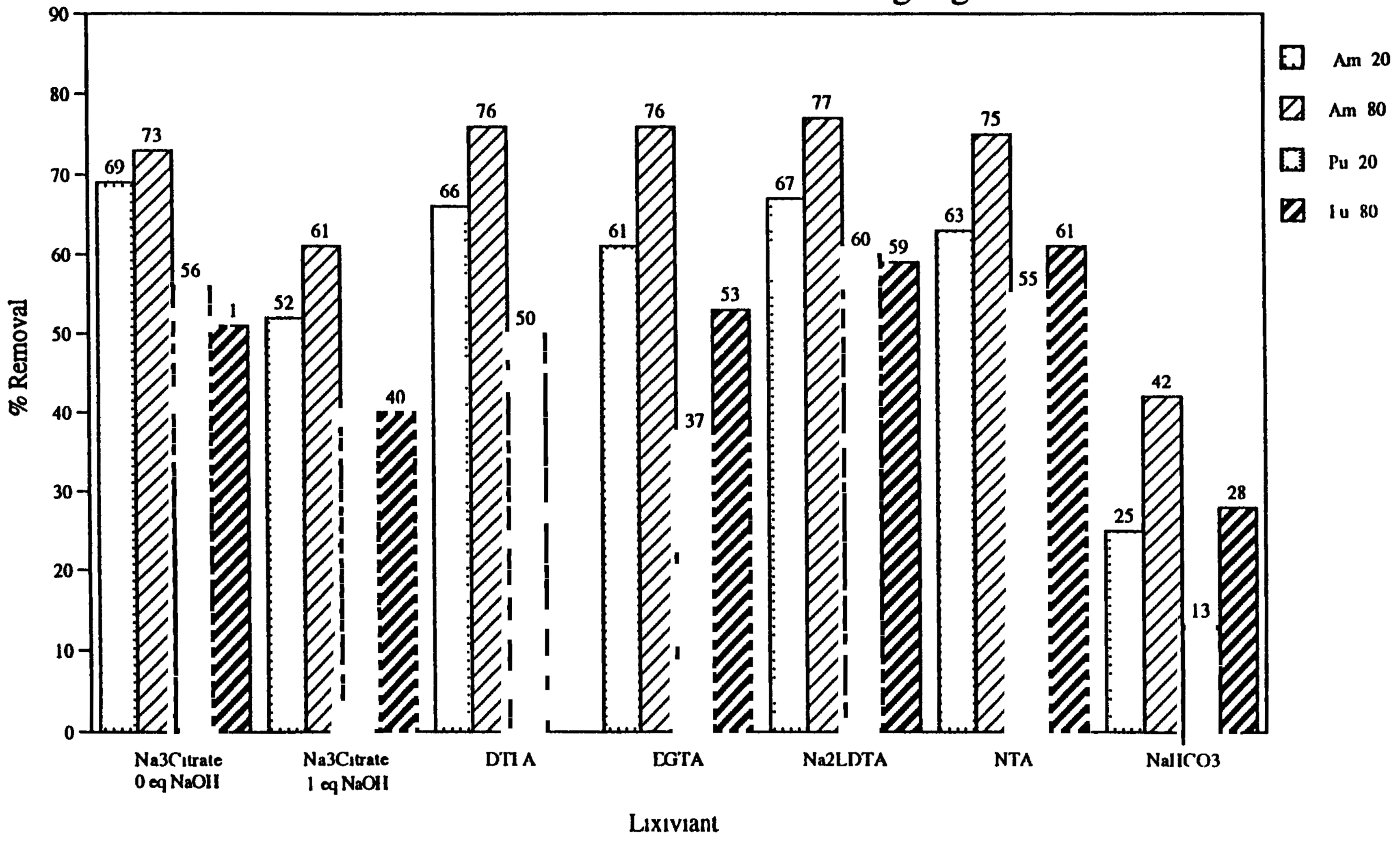


F 7

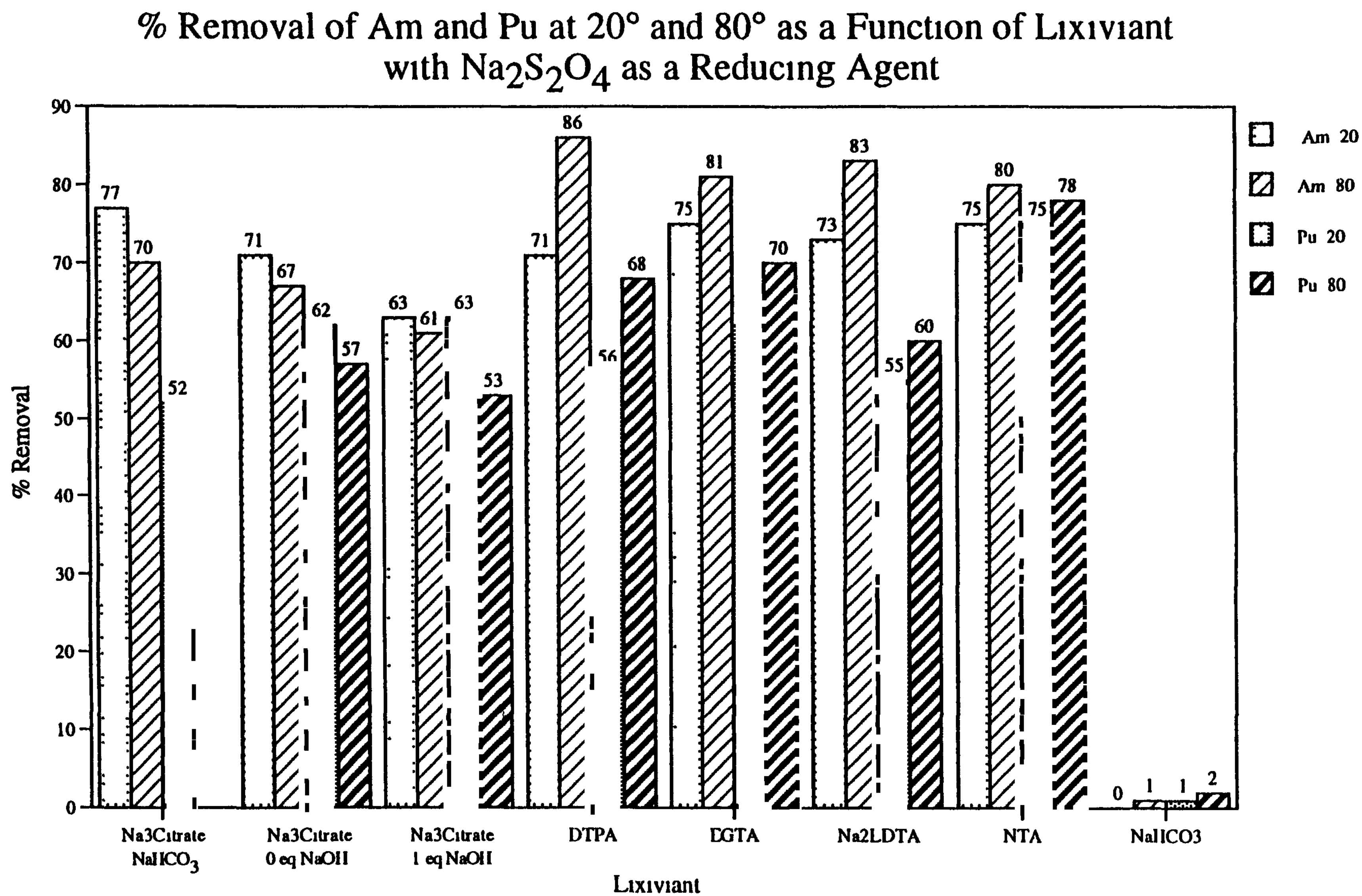
Appendix G

% Removal as a Function of Lixiviant with all Redox Agents Used at 20° and 80°

% Removal of Am and Pu at 20° and 80° as a Function of Lixiviant with Ascorbic Acid as a Reducing Agent

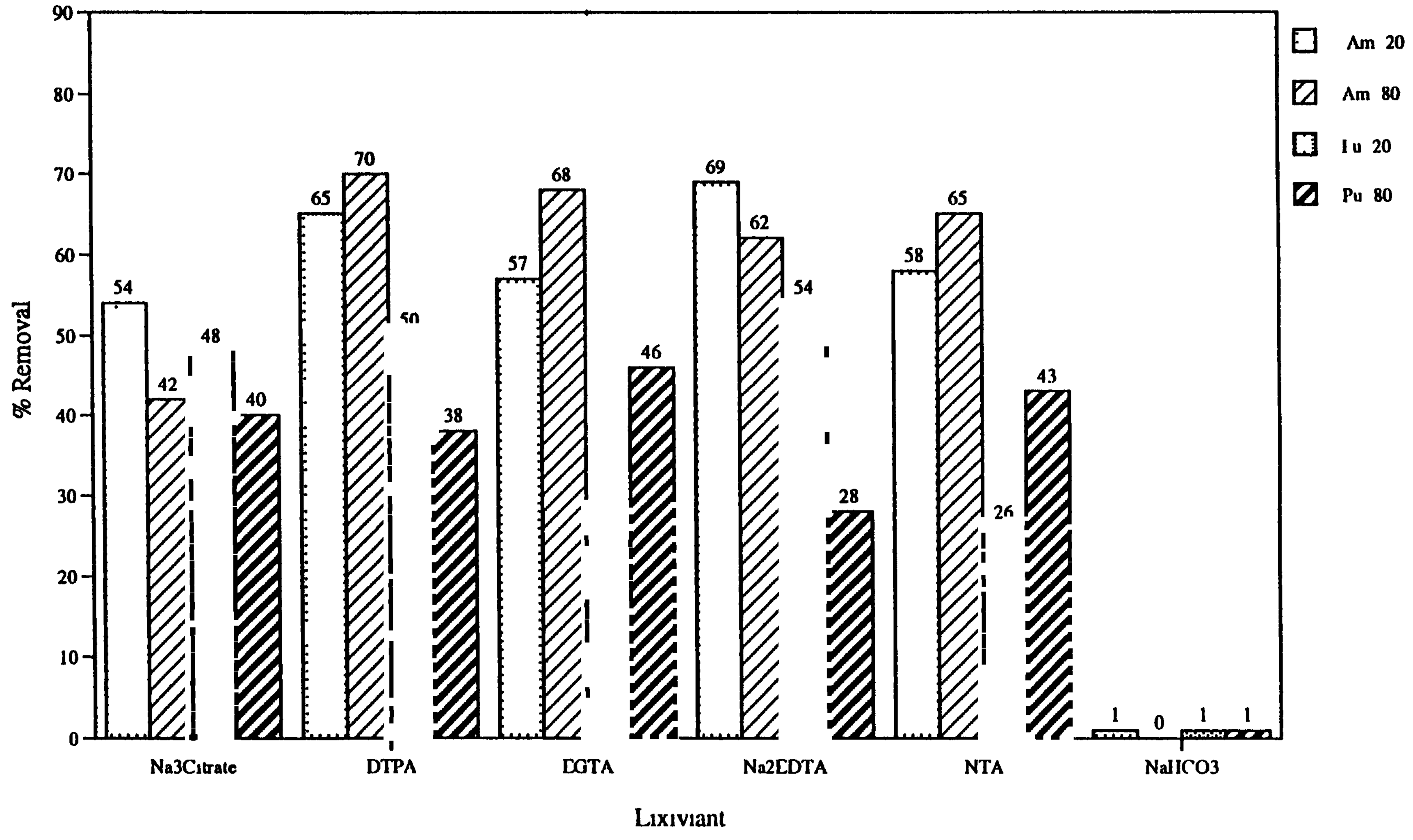


G 2


% Removal of Am and Pu at 20° and 80° as a Function of Lixiviant with $Na_2S_2O_4$ as a Reducing Agent
% Removal
0
10
20
30
40
50
60
70
80
90
77
70
52
71
67
62
57
63
61
63
53
71
86
56
68
75
81
70
73
83
55
60
75
80
75
78
0
1
1
2
Na3Citrate NaHCO3
Na3Citrate 0 eq NaOH
Na3Citrate 1 eq NaOH
DTPA
EGTA
Na2EDTA
NTA
NaHCO3
Lixiviant
Am 20
Am 80
Pu 20
Pu 80

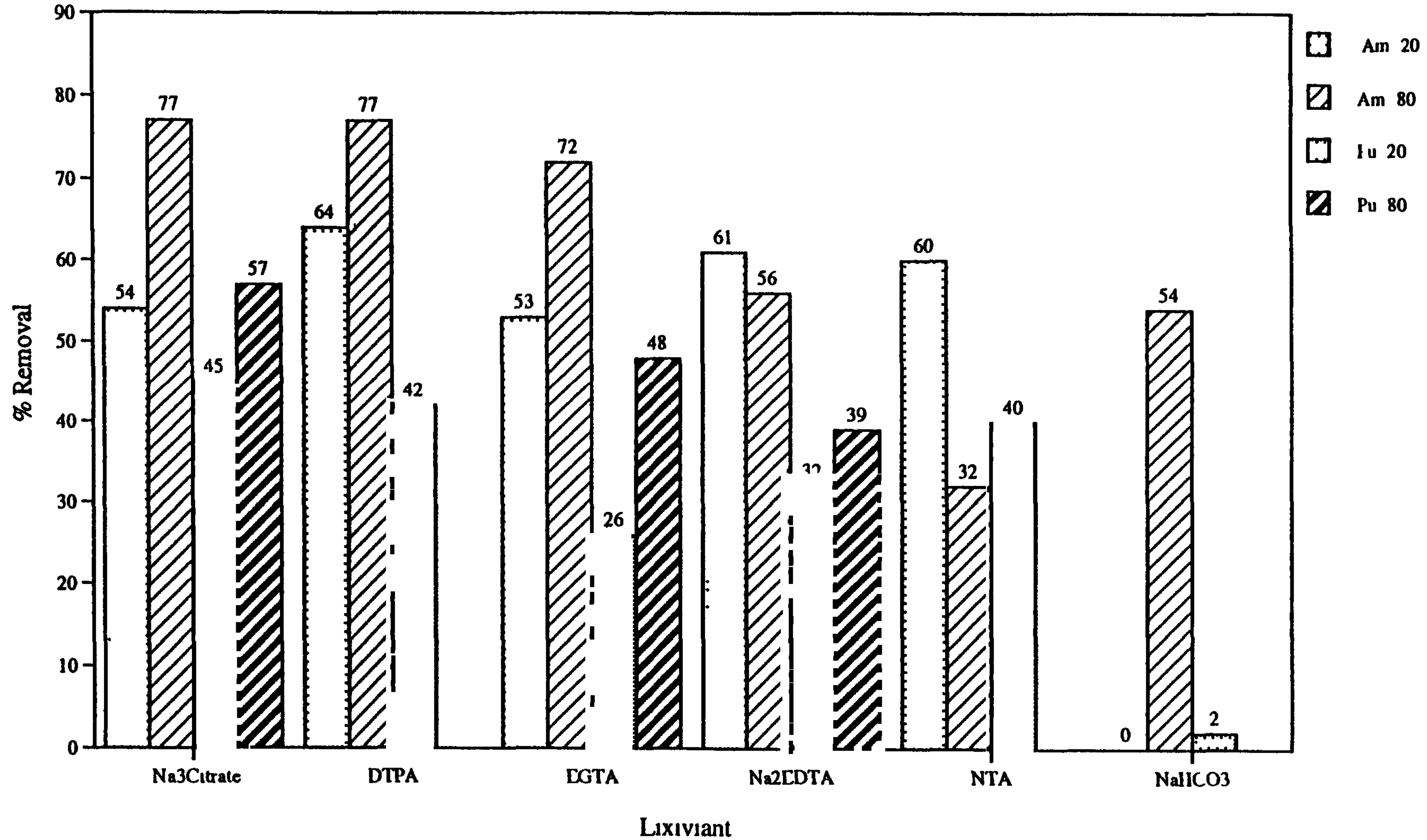
G 3

% Removal of Am and Pu at 20° and 80° as a Function of Lixiviant with 3% H_2O_2 as an Oxidizing Agent



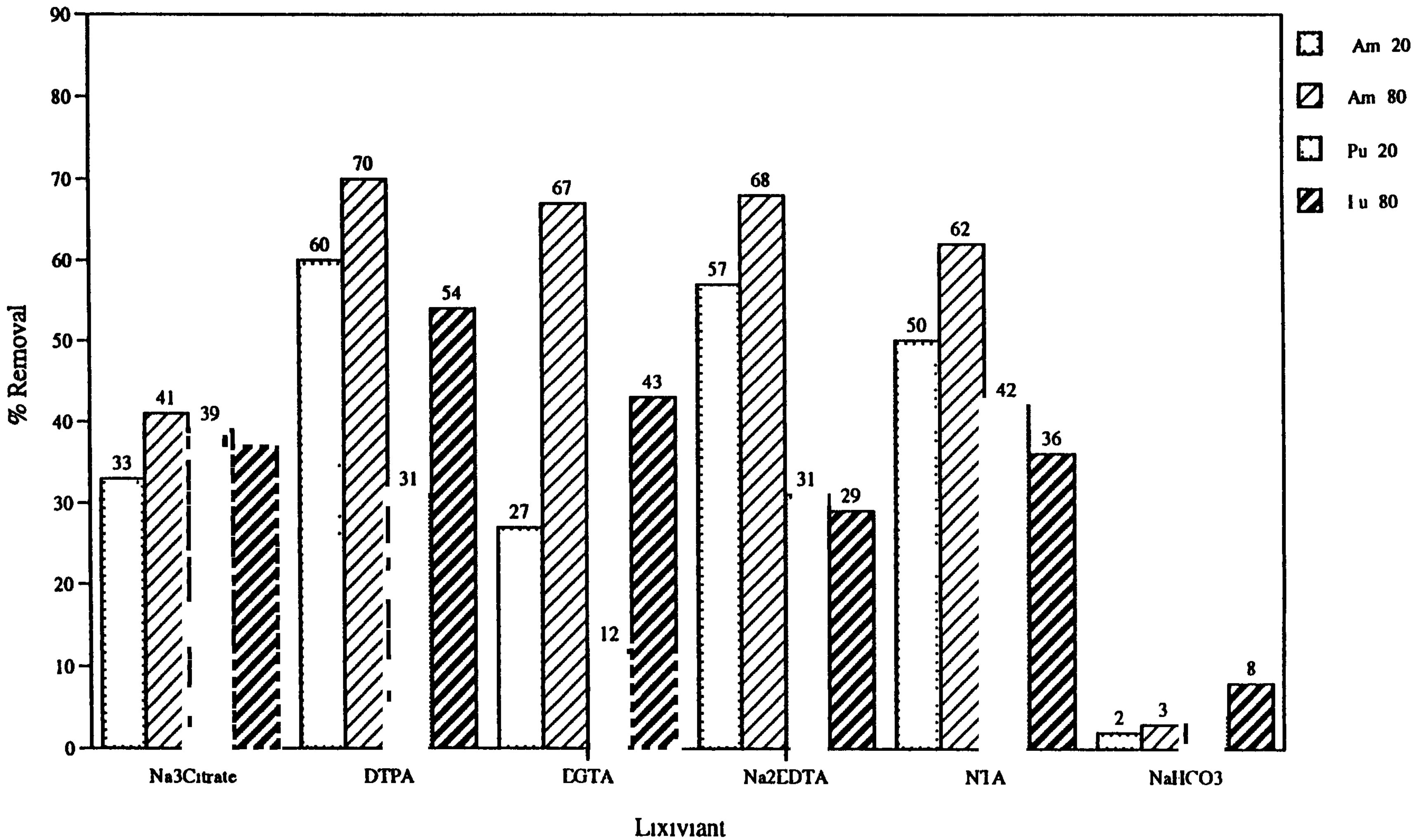
G 4

% Removal of Am and Pu at 20° and 80° as a Function of Lixiviant with $Na_2S_2O_8$ as an Oxidizing Agent



G 5

% Removal of Am and Pu at 20° and 80° as a Function of Lixiviant with 1% NaOCl as an Oxidizing Agent



G 6

Appendix H Sample Calculations

[NOTE For the 80°C desorptions the solution analyzed for its radionuclide content included 1 ml of an aqueous solution to prevent evaporation plus 5 ml of the leaching solution (after the soil was leached) This was not done for the experiments at 20° in the latter experiments 6 ml of the leaching solution after soil leaching were analyzed no extra solution was added to the bottle sent for analysis]

Let

D = Aliquot of soil leached g

E = Aliquot of leaching solution added to the soil g

L = Aliquot of leaching solution (after the soil was leached) + aqueous solution (to prevent evaporation) analyzed for its radionuclide content g

M = Aliquot of leaching solution (after the soil was leached) in liquid analyzed g

Q = Radionuclide concentration in liquid analyzed pCi/g

N = Radionuclide concentration in dry soil analyzed pCi/g

J = Liquid associated with wet soil (which was dried and analyzed for its radionuclide content after leaching) g

Rn = Radionuclide

$$\text{\% Rn removed} = \frac{\text{amt of Rn in leaching soln}}{\text{amt of Rn in leaching soln + amt of Rn in leached soil}} \times 100$$

$$\text{pCi of Rn in leaching solution (after soil leaching)} = Q \times E \times \underbrace{(L/M)}_{\text{dilution factor}}$$

$$\text{pCi of Rn in leached soil} = N \times D \quad \underbrace{[Q \times J \times (L/M)]}_{\text{this term takes into account the leaching solution that remained behind in the soil}}$$

$$\text{\% Rn removed} = \frac{Q \times E \times (L/M)}{Q \times E \times (L/M) \; + \; N \times D \quad [Q \times J \times (L/M)]} \times 100$$

For instance

% Am Removed for Sample No 296 at 80° (leaching solution Na_2EDTA second trial) =

$$\frac{45\ \text{pCi/g} \times 19\ 39\ \text{g} \times (6.25\ \text{g} / 5\ 25\ \text{g})}{45\ \text{pCi/g} \times 19\ 39\ \text{g} \times (6.25\ \text{g} / 5\ 25\ \text{g}) + 202\ \text{pCi/g} \times 2.52\ \text{g} \quad [45\ \text{pCi/g} \times 1\ 71\ \text{g} \times (6.25\ \text{g}/5\ 25\ \text{g})]} \times 100$$

$$= \frac{1038\ 8\ \text{pCi}}{1456\ 2\ \text{pCi}} \times 100$$

$$= 71\%$$

[NOTE For the 20° runs the calculation is exactly the same except that L = M and the term L/M drops out of the equation]